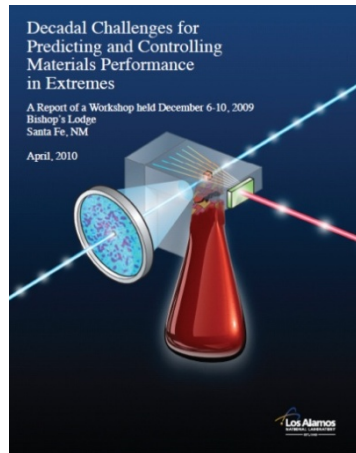


MaRIE:

(**M**atter-**R**adiation **I**nteractions in **E**xtrêmes)

An Experimental Facility Concept Revolutionizing Materials in Extrêmes

John Sarrao
Los Alamos National Laboratory





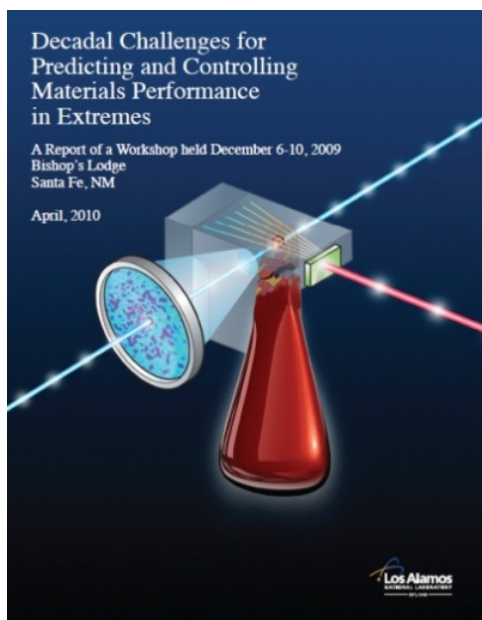
Outline

- Current Definition: The Why and What of MaRIE
 - *Why would you want to build a 50 keV XFEL and couple it to a MW-class proton accelerator?*
- Acquisition Strategy and Facility Realization
 - *Why might Los Alamos be an interesting place to do this?*
- Where we need your help
 - *How might you become involved, especially in advance of the first 'call for proposals' in ~ 2020?*



Materials research is on the brink of a new era – moving from observation of performance to control of properties

- The confluence of improved experimental capabilities (e.g. 4th generation light sources, controlled synthesis and characterization, ...) and simulation advances are providing remarkable insights at length and time scales previously inaccessible



New capabilities will be needed to realize this vision:

In situ, dynamic measurements

simultaneous scattering & imaging

of well-controlled and characterized materials

advanced synthesis and characterization

in extreme environments

dynamic loading, irradiation

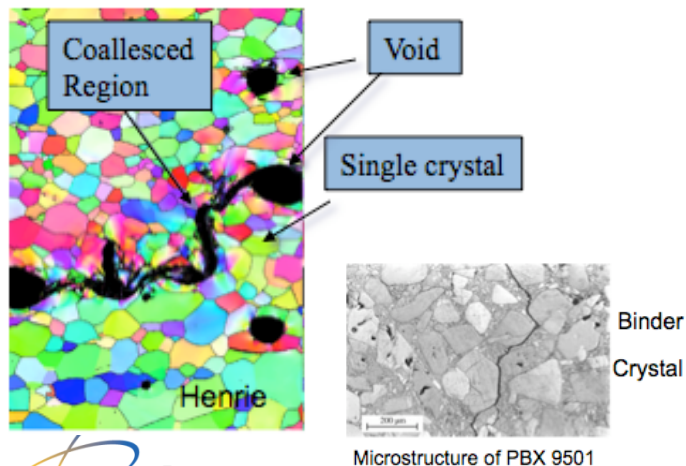
coupled with predictive modeling and simulation

materials design & discovery

Understanding materials in extreme environments is key to weapons program success in the future



MATERIALS MATTER



Operated by Los Alamos National Security, LLC for NNSA

Current Stockpile

- Prediction of materials lifetime & failure

Rebuild & Lifetime Extension

- Materials 'by design' rather than re-learning old processes

Weapon performance

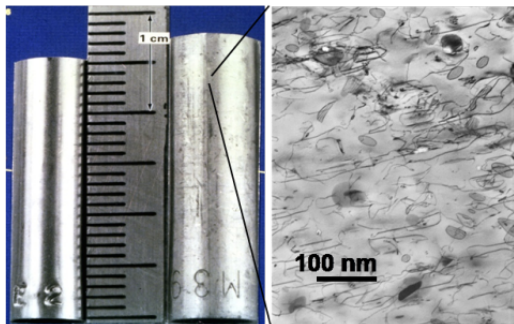
- Effects of microscale materials properties on dynamic performance for key physics

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Slide 4



Materials behavior limits the performance of advanced energy systems needed for energy independence

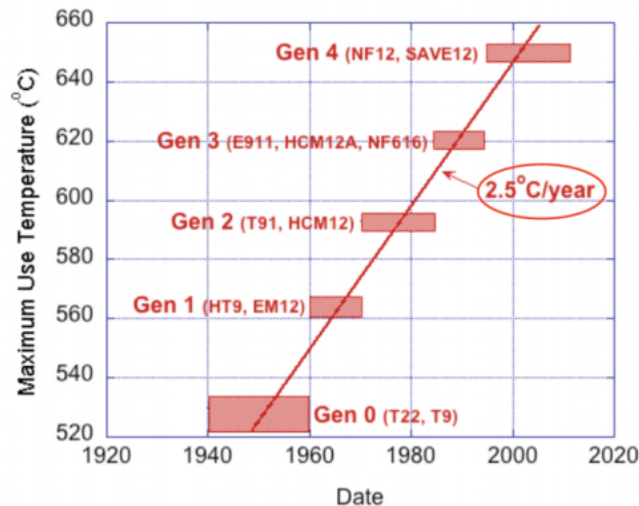


Life extension, safety of existing reactor fleet

Improved affordability for new reactors

Sustainable fuel cycles

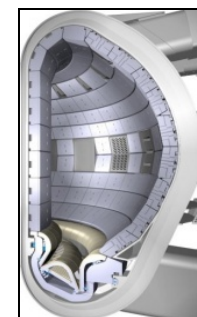
Fusion Reactor first wall materials



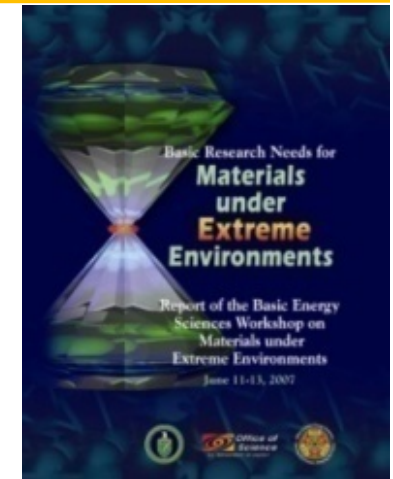
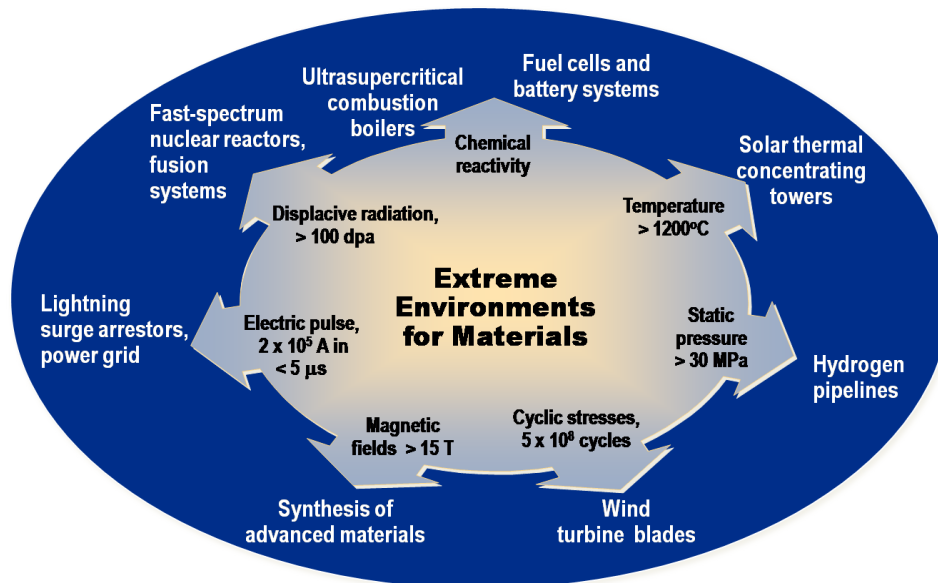
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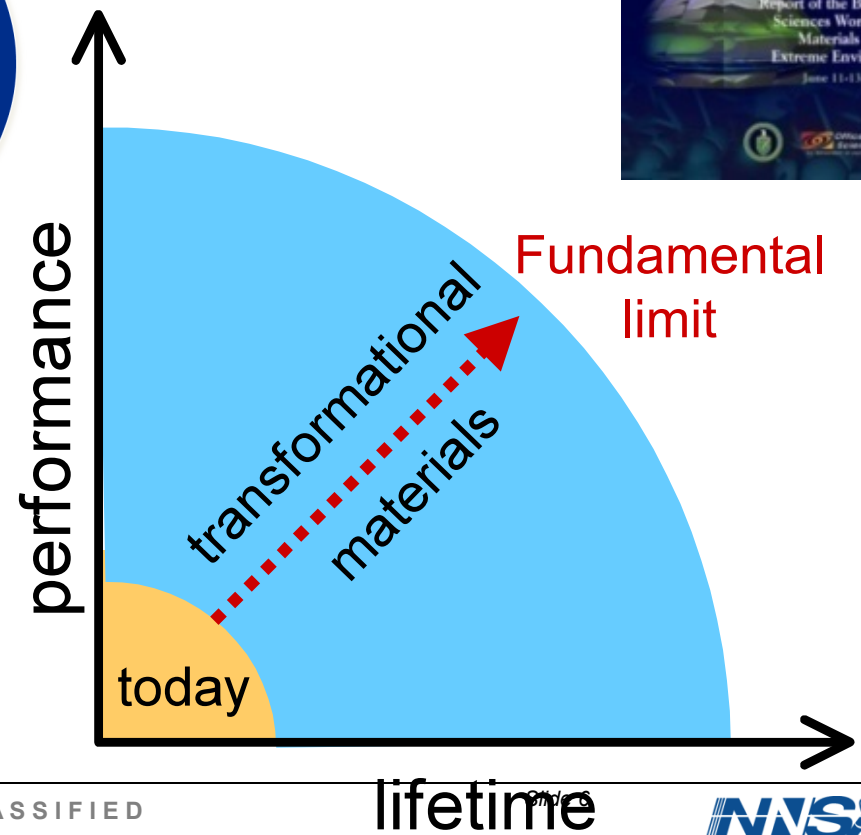
Slide 5



The needs for materials in extremes are many; the challenge is common: revolutionary advances in controlled functionality



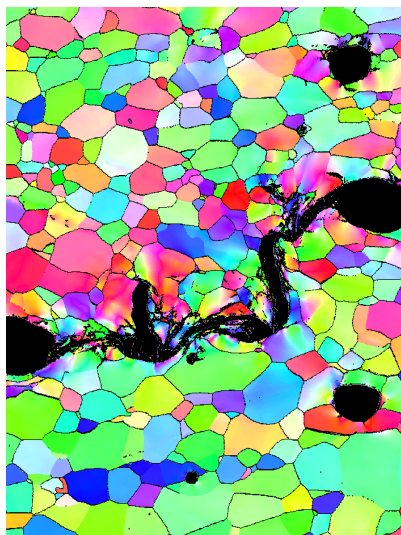
We need to enable a transition:
from observation and validation
to prediction and control



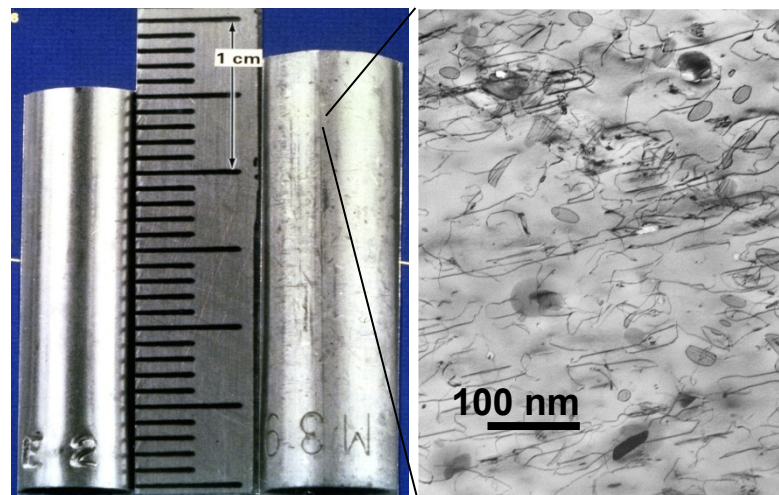


The “micron frontier:” Bridging the gap between atomic understanding and bulk performance

~ 1 μm is the domain of defect consequences and microstructure interactions that drive materials strength, damage evolution, etc.



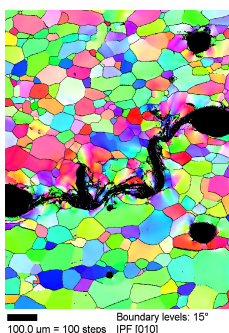
100.0 μm = 100 steps
Boundary levels: 15°
IPF [010]



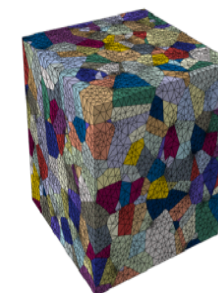
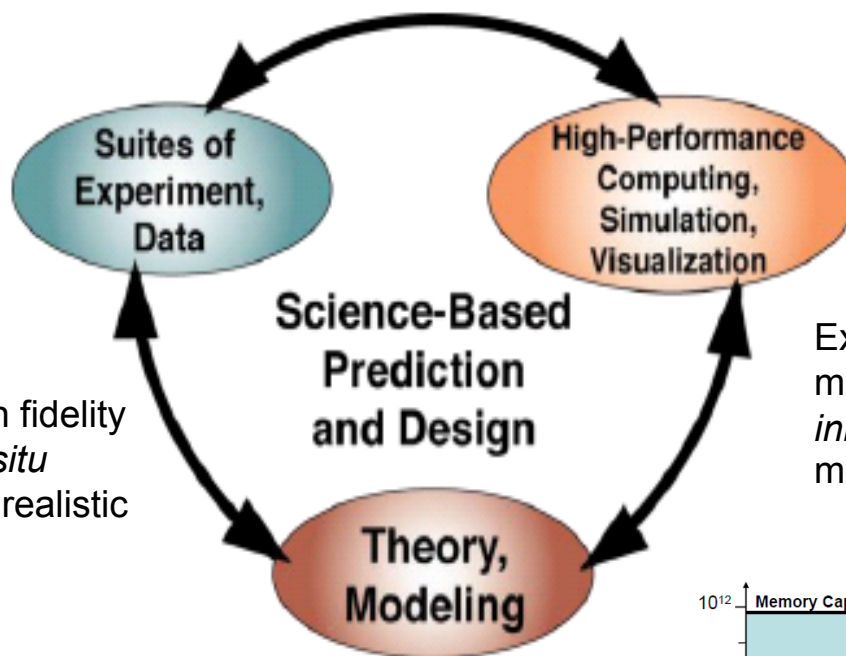
Dynamic, stochastic processes in extreme environments dominate the phenomena that we do not understand



Next generation simulation capabilities and experimental tools will enable discovery science at the micron frontier

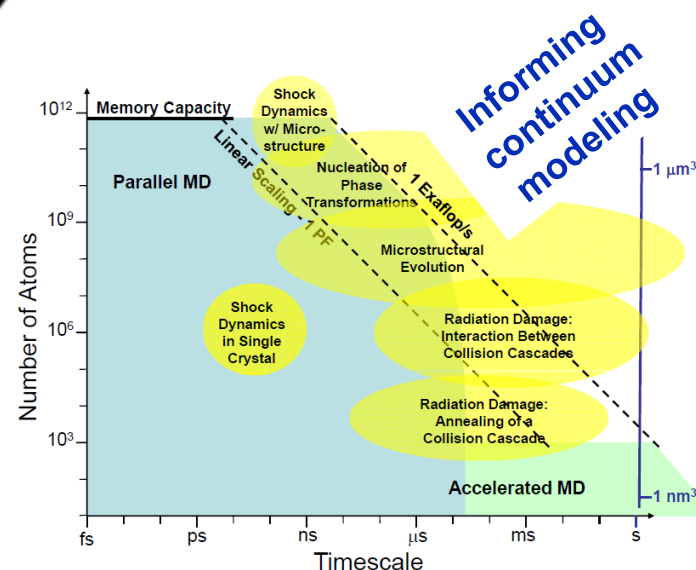


Controlled fabrication, high fidelity characterization, novel *in situ* diagnostics, generation of realistic extreme environments, ...

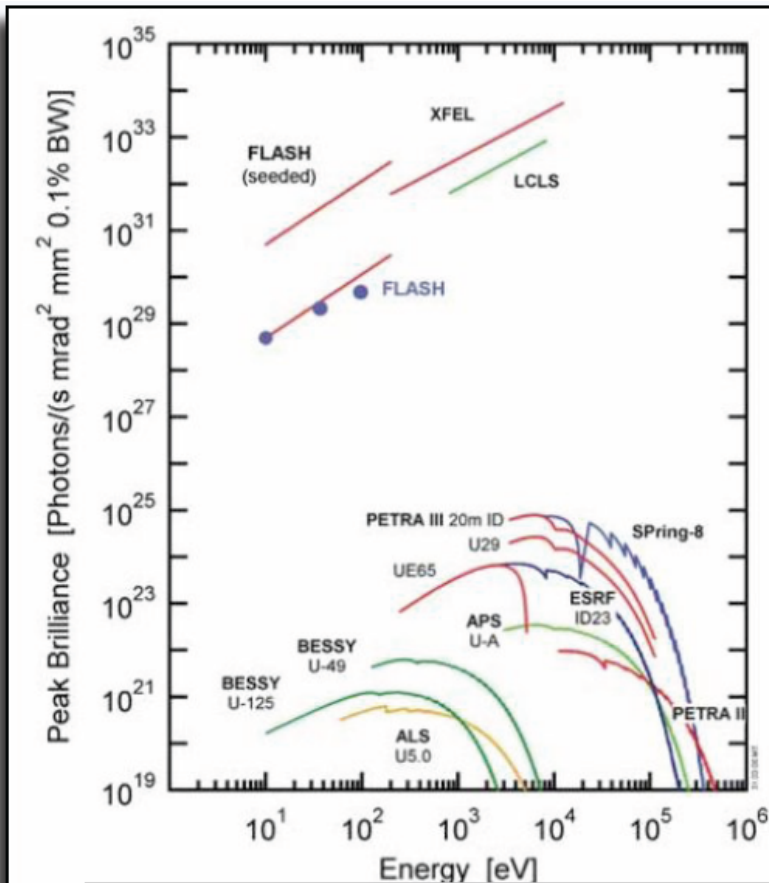


Exascale computing, multi-scale, multi-physics simulation tools, *ab initio* methods applied to larger, more complex materials, .

Multi-scale approaches to connect fundamental scales to bulk properties, defect generation and evolution, ...



4th generation light sources, producing brilliant, coherent photons, provide a unique opportunity (especially when coupled with broader, integrated capabilities)



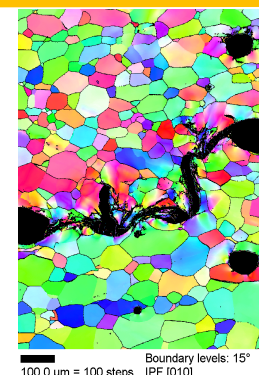
	FLASH (6.0 nm)	LCLS (0.15 nm)	XFEL (0.1 nm)
mJ/pulse	0.3	2.6	3.7
Photons/pulse	9×10^{12}	2×10^{12}	2×10^{13}
GW	3	26	37
Peak Brightness	2.0×10^{30}	1.2×10^{33}	8.7×10^{33}
Bandwidth (%)	0.6	0.3	0.1
Hz	50	100	50
Date	2005	2009	2015

100 fs time resolution, sub-nm spatial resolution, high peak power ($>10^{17}$ W/cm²), full transverse coherence

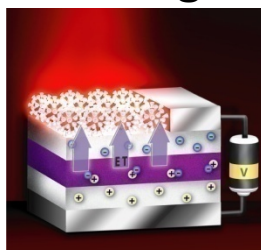


MaRIE : What does success look like?

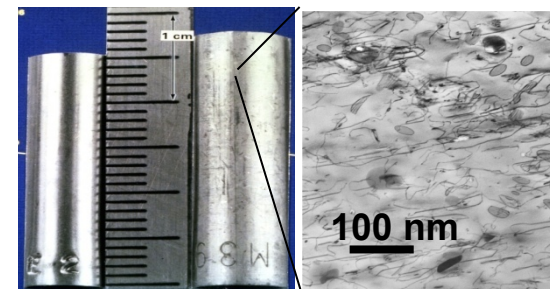
- Predicting materials performance, including failure, in extremes of pressure and strain for multi-phase materials
- Developing radiation resistant structural materials and fuels by design
- Exploiting complex materials and architectures for next generation electronics



Materials failure under dynamic load



Next-generation solar cell architecture



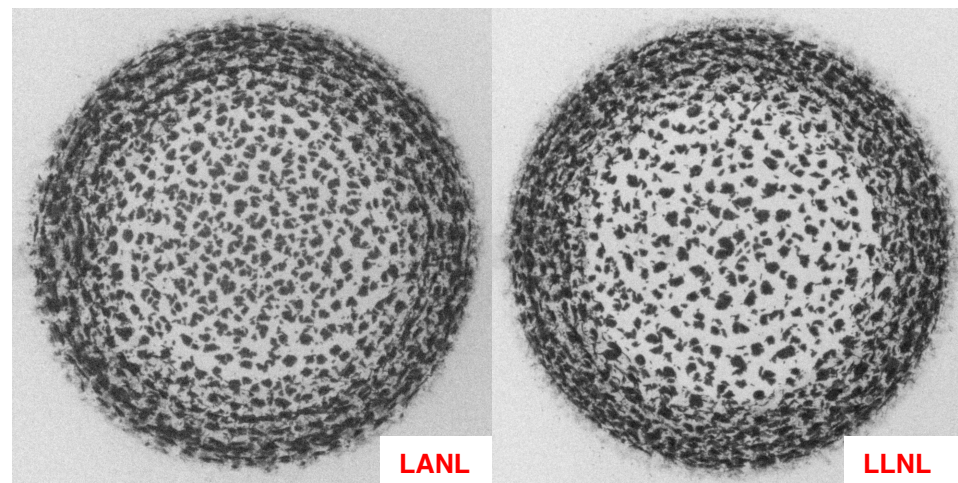
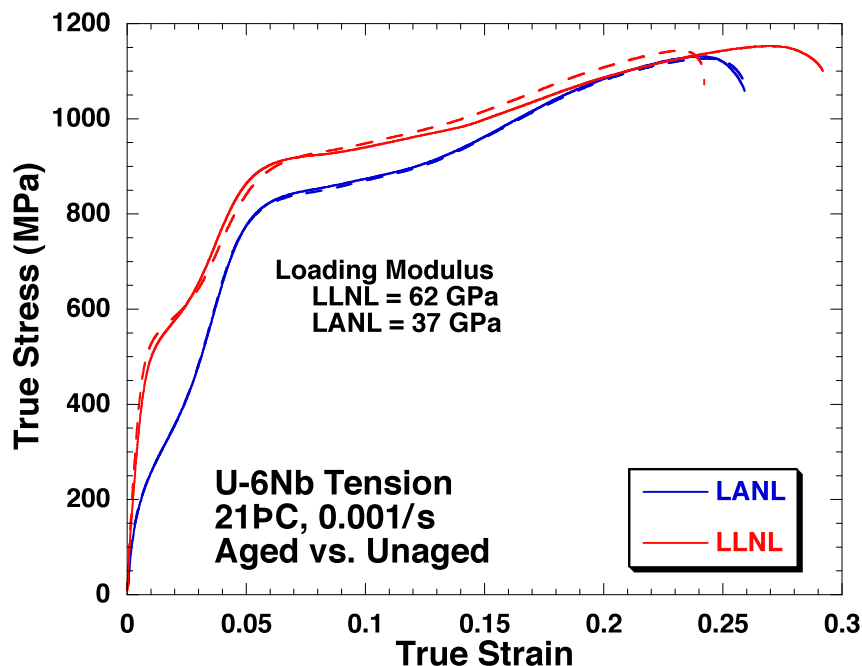
Radiation-induced swelling



Mechanical behavior and HE-driven fragmentation of U-6%Nb show strong influence of metallurgical state

LANL = Solution treated / Quenched

LLNL = Solution treated / Quenched + Aged

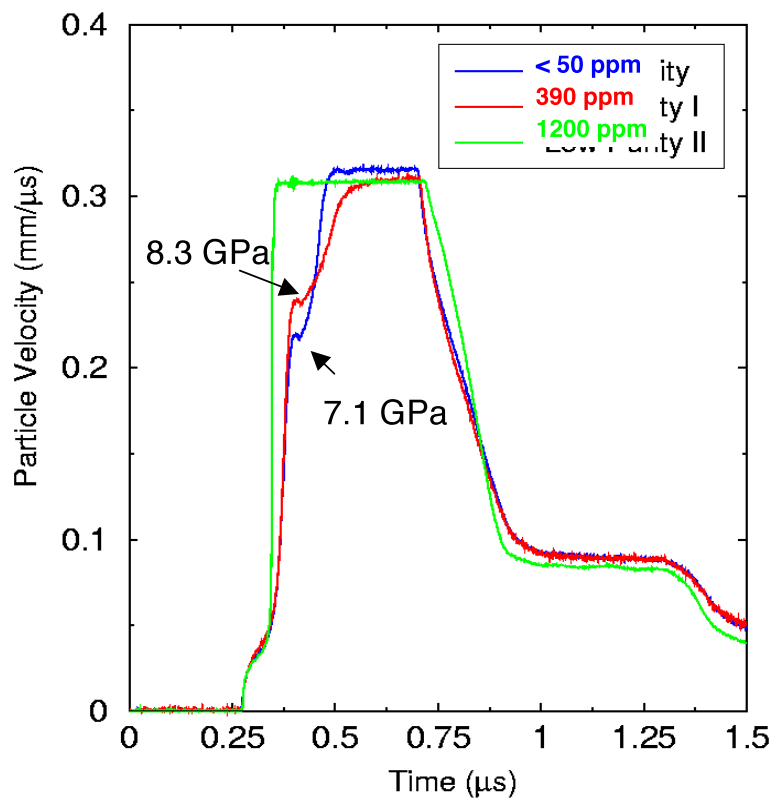


Quantitative analysis reveals 6σ difference in open area between images



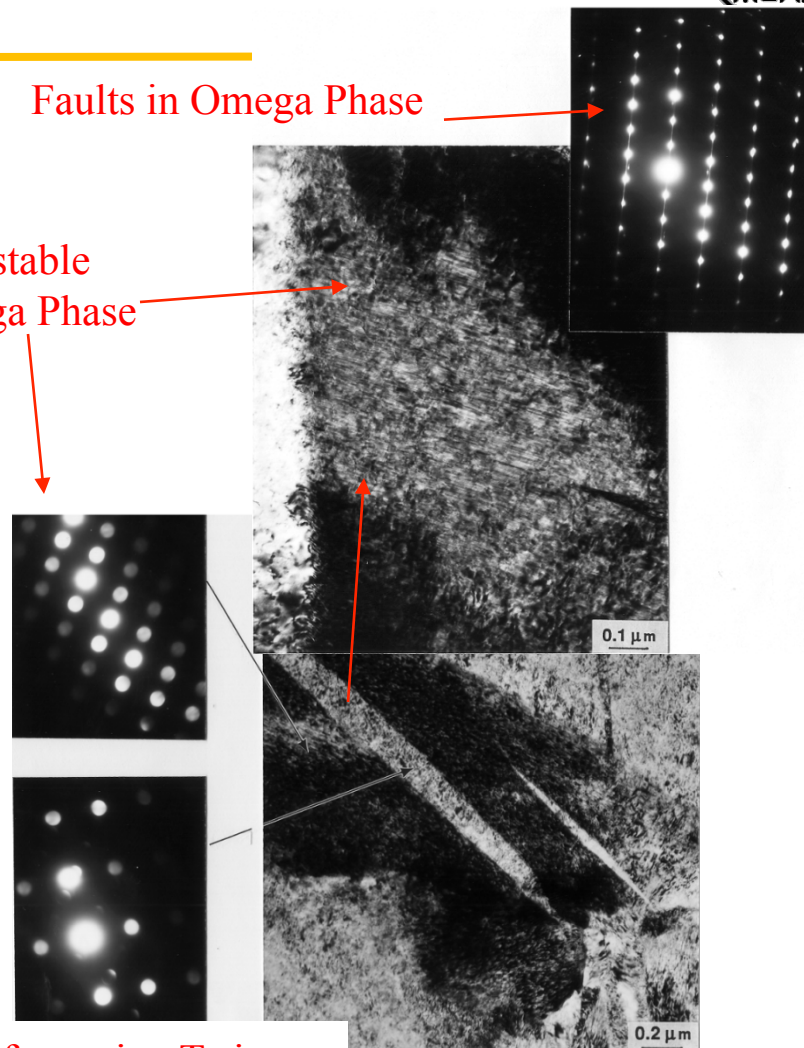
Shock-induced phase transitions reveal spatially complex processes with strong materials sensitivities

Oxygen content suppresses the α - ω phase transition in Zr



Faults in Omega Phase

Metastable Omega Phase

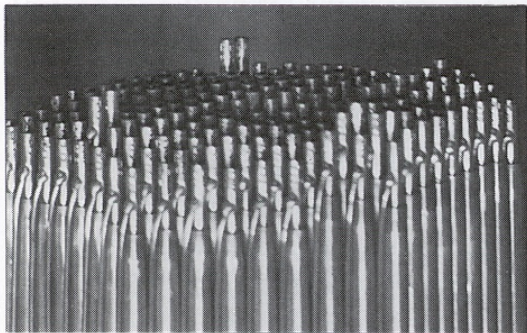


Deformation Twins

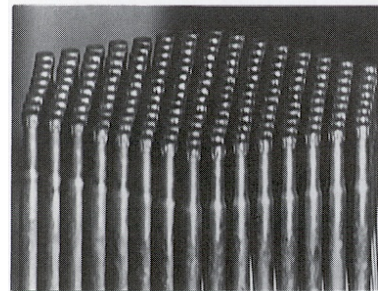
Radiation-resistant materials have tended to be developed serendipitously and empirically



- Ferritic/martensitic steels (like HT9) are leading candidates for cladding, structural materials of fast breeder reactors (FBRs) and the first walls and blankets in conceptual fusion reactor designs
- They show resistance to void swelling and have adequate mechanical properties at elevated temperatures → expanded operating environments

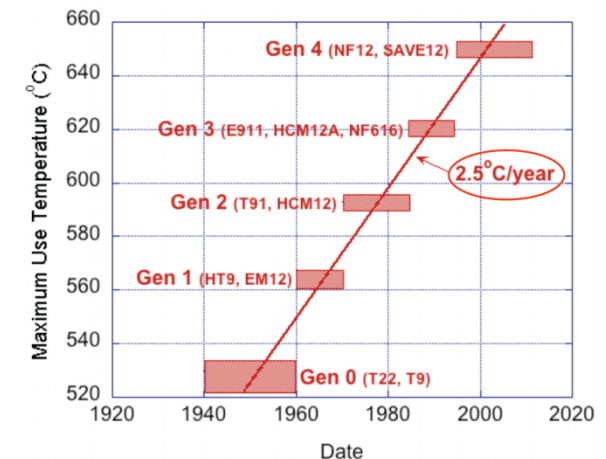


D9 irradiated to $2.1 \cdot 10^{23}$ ($E > 0.1 \text{ MeV}$)*



HT9 irradiated to $1.9 \cdot 10^{23}$ ($E > 0.1 \text{ MeV}$)*

* Makenas et al 1990

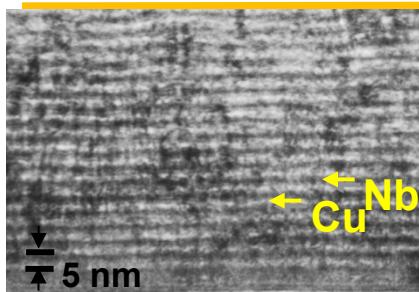


after Zinkle, Busby Mater. Today 12 (2009) 12

However, our understanding of the atomic-level processes that control bulk behavior is substantially incomplete

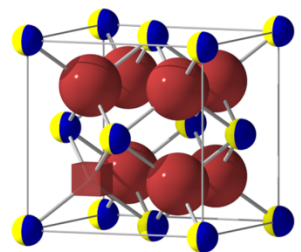
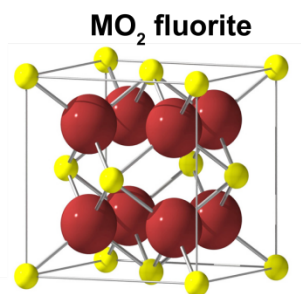
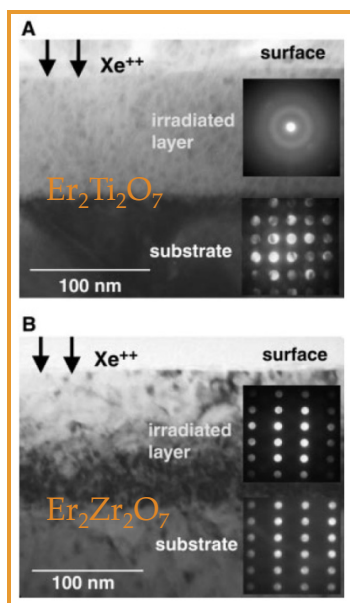


Frontiers of materials discovery: Interface/structure manipulation produces enhanced strength and radiation resistance

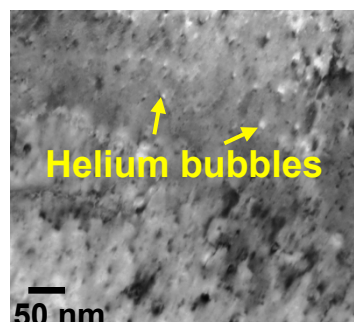


Nanolayer architectures produce materials strength that exceeds theoretical “limits”

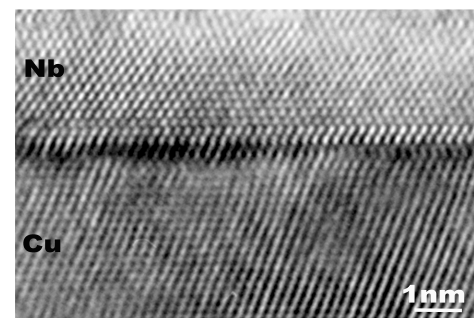
Same structures produce extreme radiation resistance by actively eliminating point defects



MO_{2-x} fluorite derivative



Pure Cu

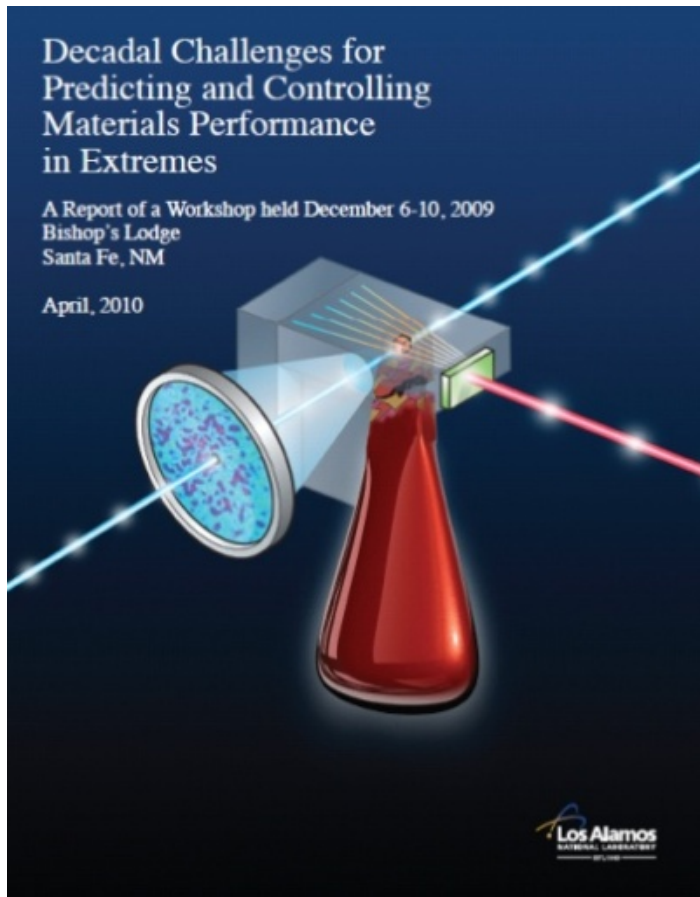


5 nm layer thickness Cu-Nb multilayer

← Challenge is to translate these insights to bulk systems

Can we discover ‘by design’ bulk materials that embody these principles and observe and manipulate their defect structures in ‘real’ deformation and irradiation extremes?

Materials research is on the brink of a new era of science in which the traditional approach of observation & validation of performance is replaced by prediction & control of materials functionality



MaRIE builds on unique LANL capabilities to provide unique experimental tools needed to realize this vision:

In situ, dynamic measurements of real materials

Scattering & imaging simultaneously

in extreme environments

Dynamic & irradiation extremes

coupled to directed synthesis via predictive theory

Materials design & discovery



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MaRIE builds on the LANSCE facility to provide unique experimental tools to meet this need

First x-ray scattering capability at high energy and high repetition frequency with simultaneous charged particle dynamic imaging

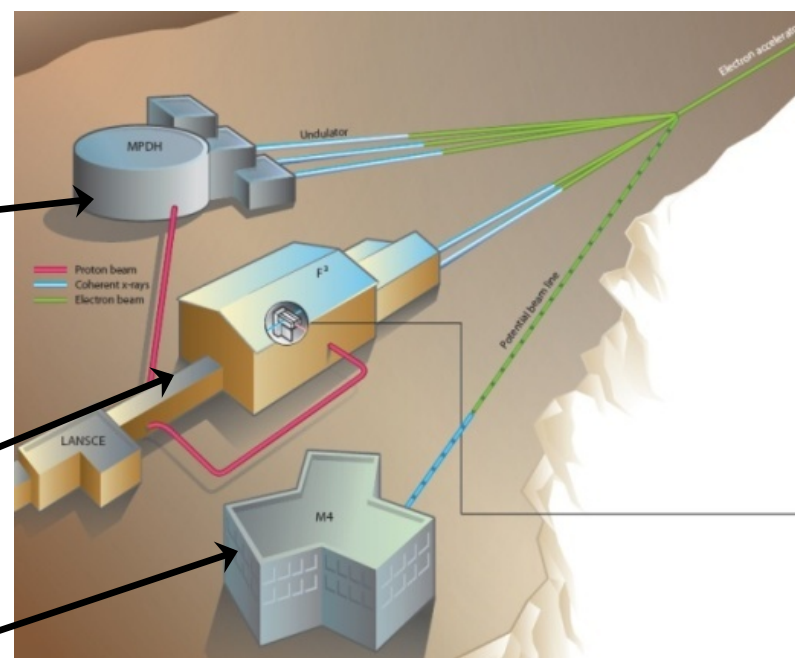
(MPDH: Multi-Probe Diagnostic Hall)

Unique in-situ diagnostics and irradiation environments beyond best planned facilities

(F³: Fission and Fusion Materials Facility)

Comprehensive, integrated resource for materials synthesis and control, with national security infrastructure

(M4: Making, Measuring & Modeling Materials Facility)



Unique very hard x-ray XFEL

Unique simultaneous photon-proton imaging measurements

Unique spallation neutron-based irradiation capability

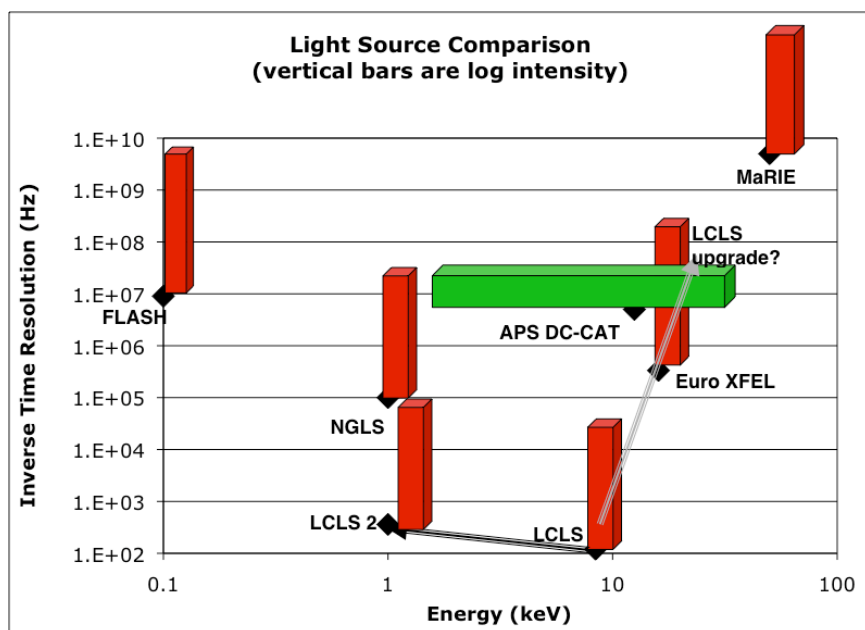
Unique in-situ, transient radiation damage measurements

Unique materials design and discovery capability

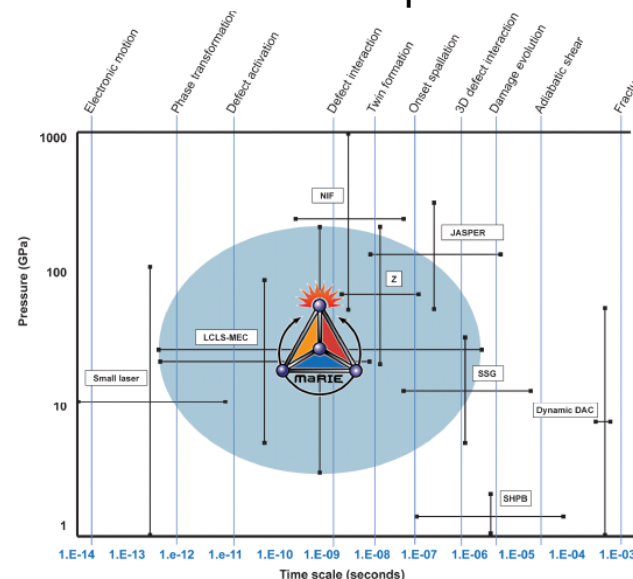


Through Multi-Probe Diagnostic Hall, MaRIE provides unique scattering and imaging capabilities to bridge the micron gap in extreme environments

A high-energy-photon (50-115 keV) XFEL allows multigranular sample penetration and multipulse dynamics without significant sample perturbation

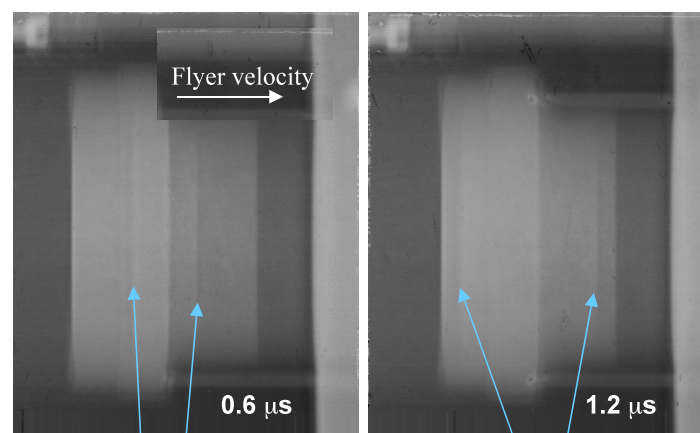


At intermediate pressures



Meanwhile, proton microscopy can provide absolute density & velocities through the sample volume

(pRad absolute Density: ~1%)

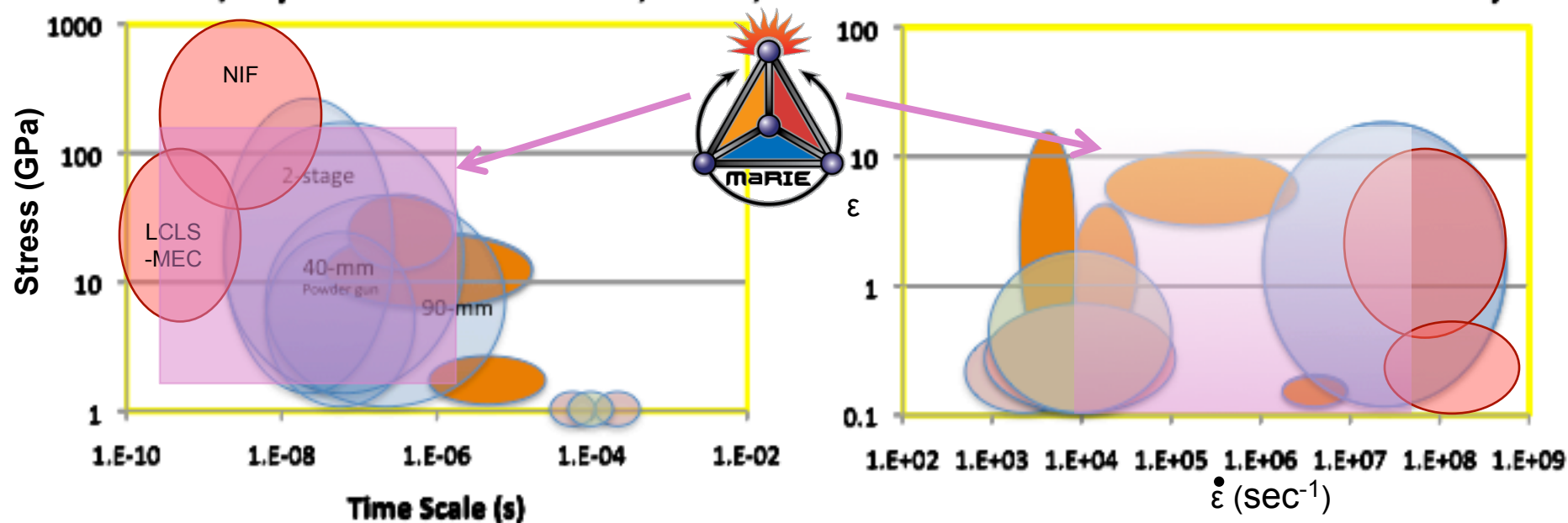


Slide 17



We are further evaluating scientific requirements on dynamic (and static) extremes needed for the science

Pressure, experimental time scale, strain, and strain rates accessible with this technique:



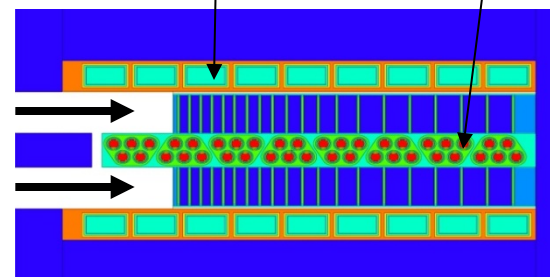
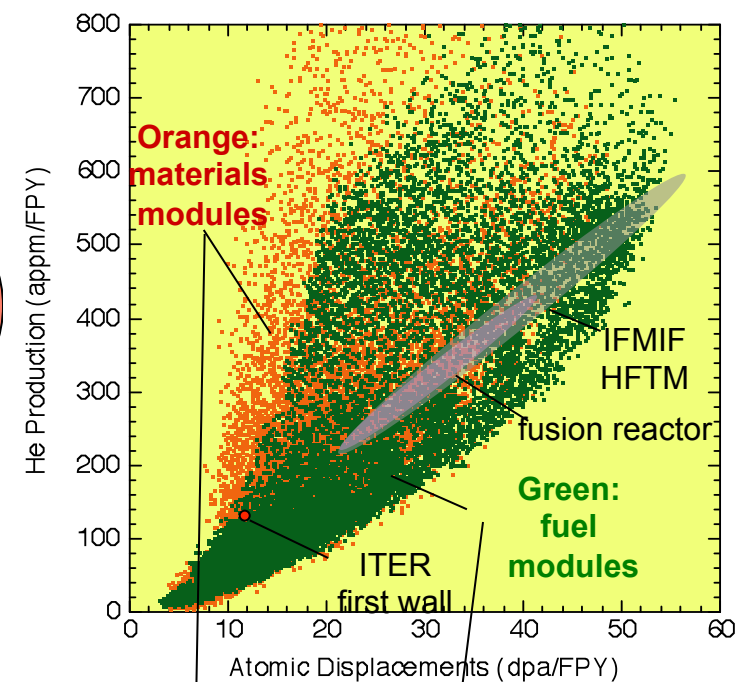
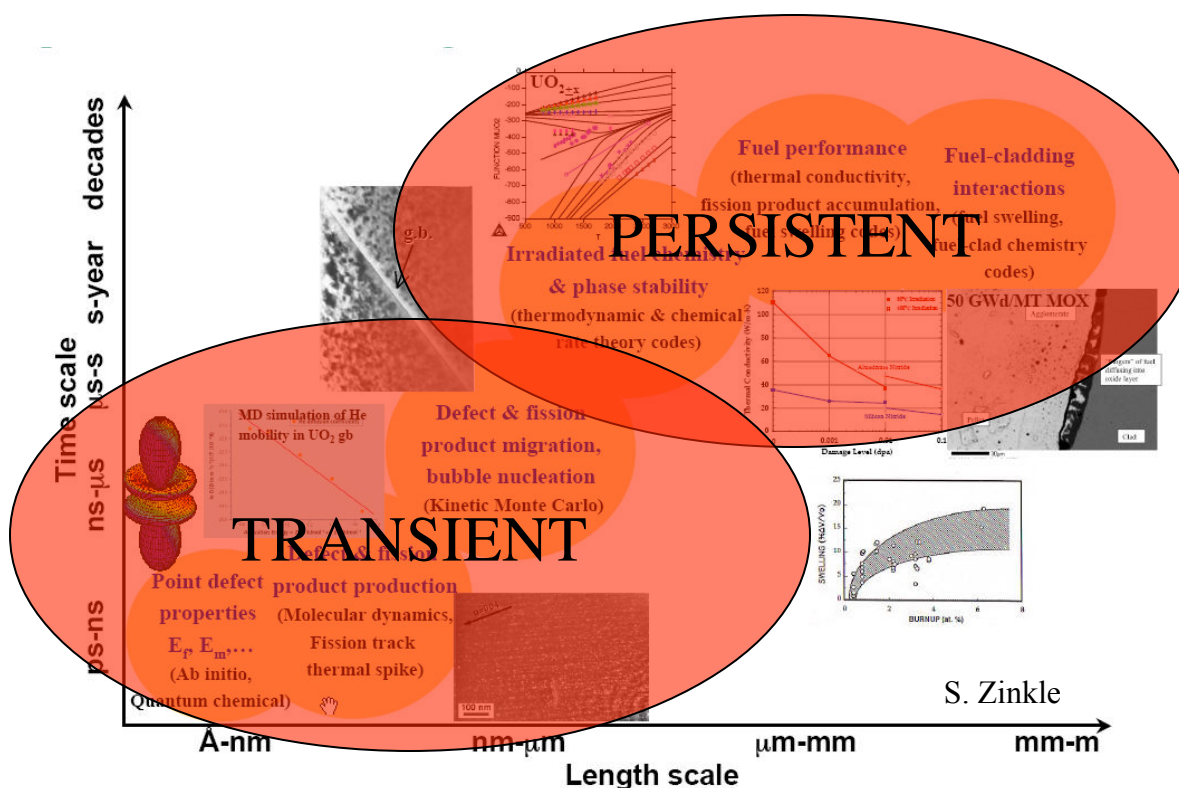
Summary developed by Science Campaign 2 on stress, time, and strain and strain rate regimes of present techniques



Through Fission Fusion Materials Facility, MaRIE creates extreme radiation fluxes and advances the frontiers of radiation damage science through in situ measurements

The same x-rays (protons) enable in-situ (near in-situ) measurements...

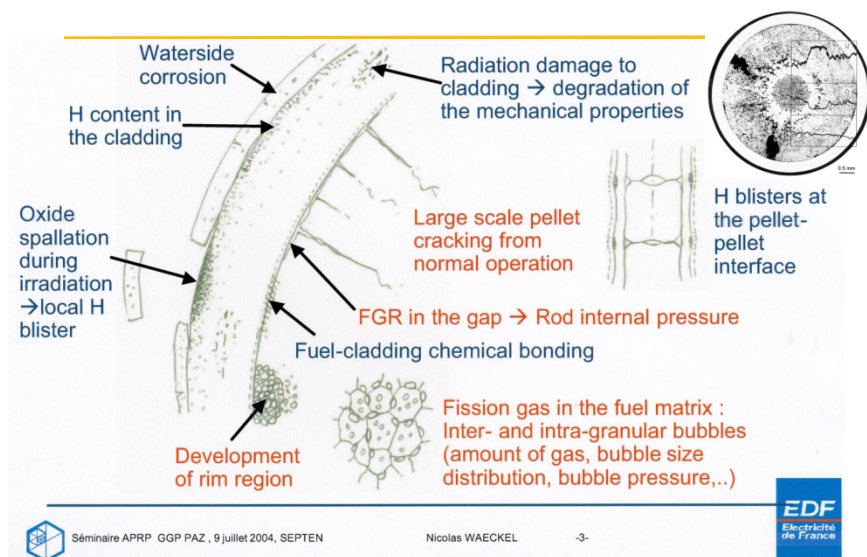
...in relevant environments



MTS target assembly



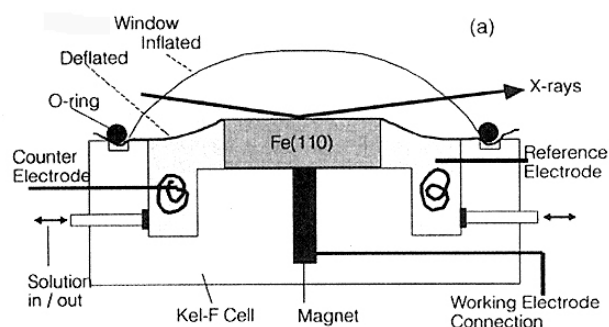
For **some** key phenomena, *in situ measurements* in a neutron environment are essential for achieving a predictive understanding



For example,

- Transient aspects of irradiation assisted stress corrosion cracking
- Corrosion under different coolants, higher temperatures and fluxes
- Transient changes of micro-structure and stress under irradiation (e.g. creep)
- Custom tests for model validation
- Transient safety tests
- Short duration low-burn up data
- Active temperature control tests

Monitor changes in fuels



Monitor corrosion conditions



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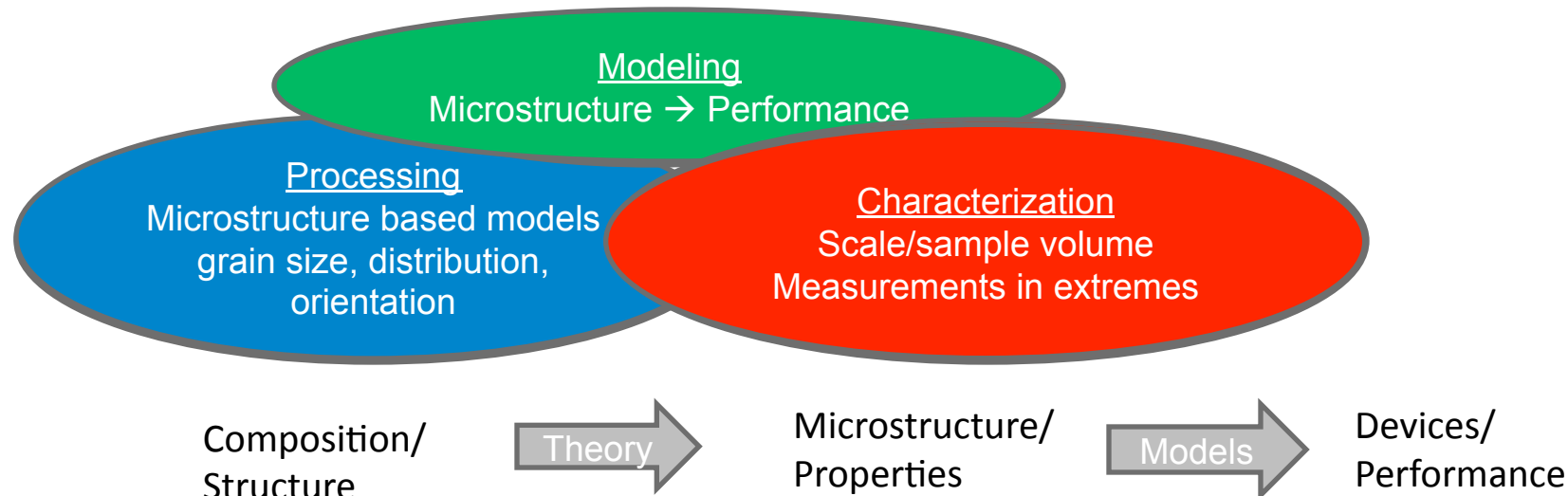
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Making, Measuring, Modeling Materials (M4): Accelerating complex materials design and discovery requires integration

Process Aware Materials Performance



“Physicists perform elegant experiments on crummy samples while materials scientists perform crummy experiments on elegant samples”

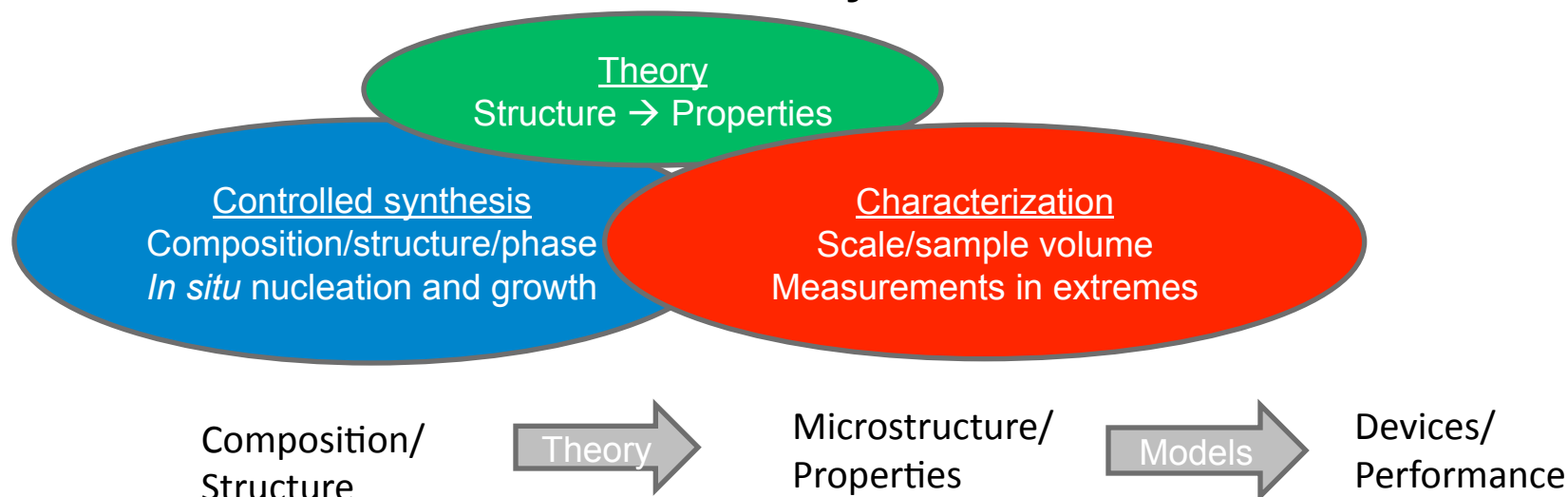
-Sig Hecker
Former LANL Director
(materials scientist)



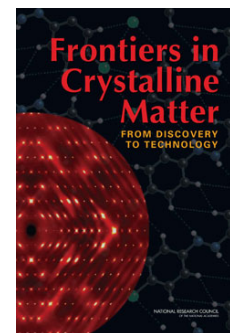


Making, Measuring, Modeling Materials (M4): Accelerating complex materials design and discovery requires integration

Materials Discovery

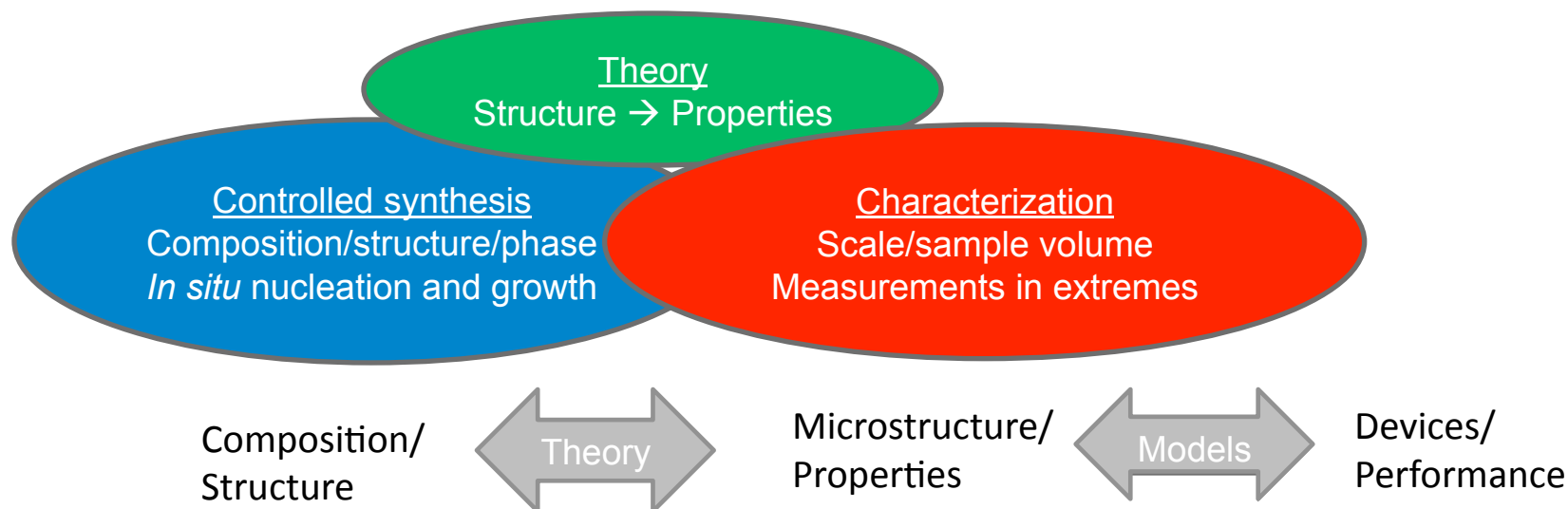


“A systematic, highly coordinated research effort in which synthesis is strongly coordinated with modeling and the characterization of novel materials with controlled ... structures, tailored surface functionality, and nanostructured architectures is critically needed”

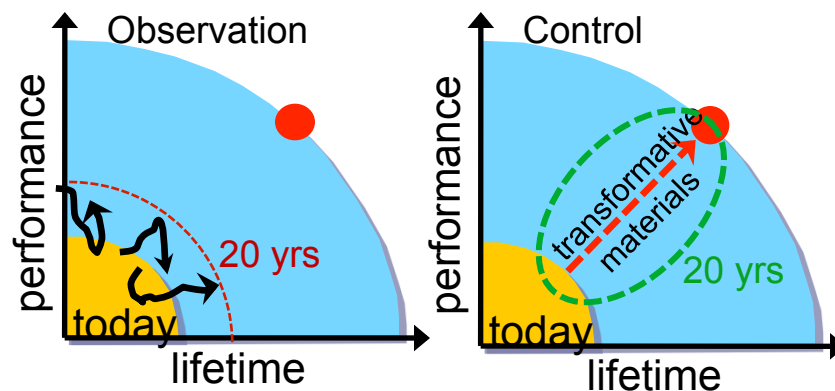




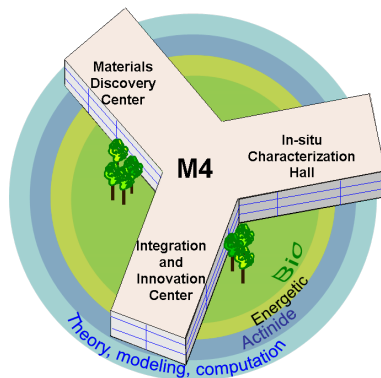
Making, Measuring, Modeling Materials (M4): Accelerating complex materials design and discovery requires integration



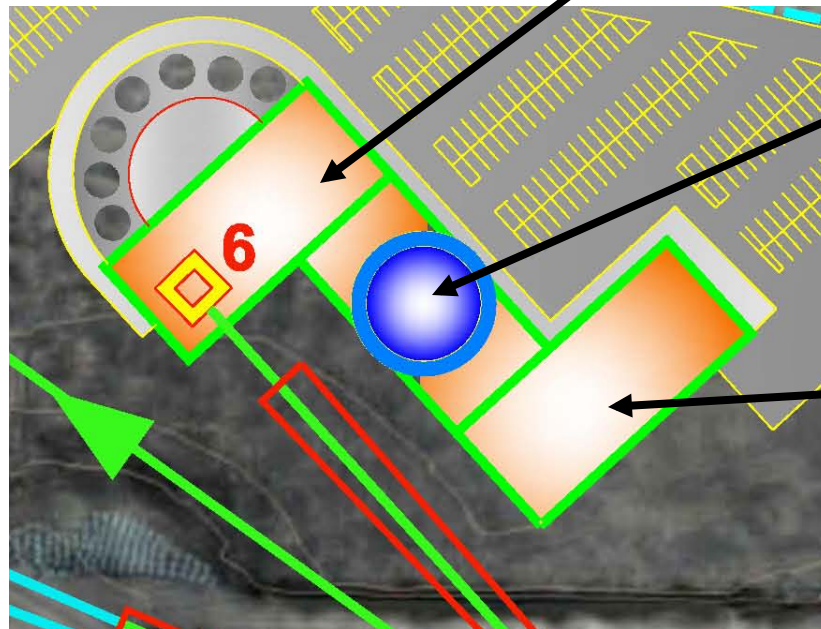
We must move from
structure → property paradigm
To a new paradigm of
function → structure



Through M4 Facility, MaRIE provides the directed synthesis of materials essential for defect/interface control and materials discovery



M4 XFEL end station
Other Extremes (E,H, pH)
In situ synthesis probes



User Gateway
Co-design Center
Visualization Capability

Multi-scale Synthesis & Crystal Growth
Characterization
National Security Infrastructure

Measuring

Modeling

Making

LOS ALAMOS
NATIONAL LABORATORY
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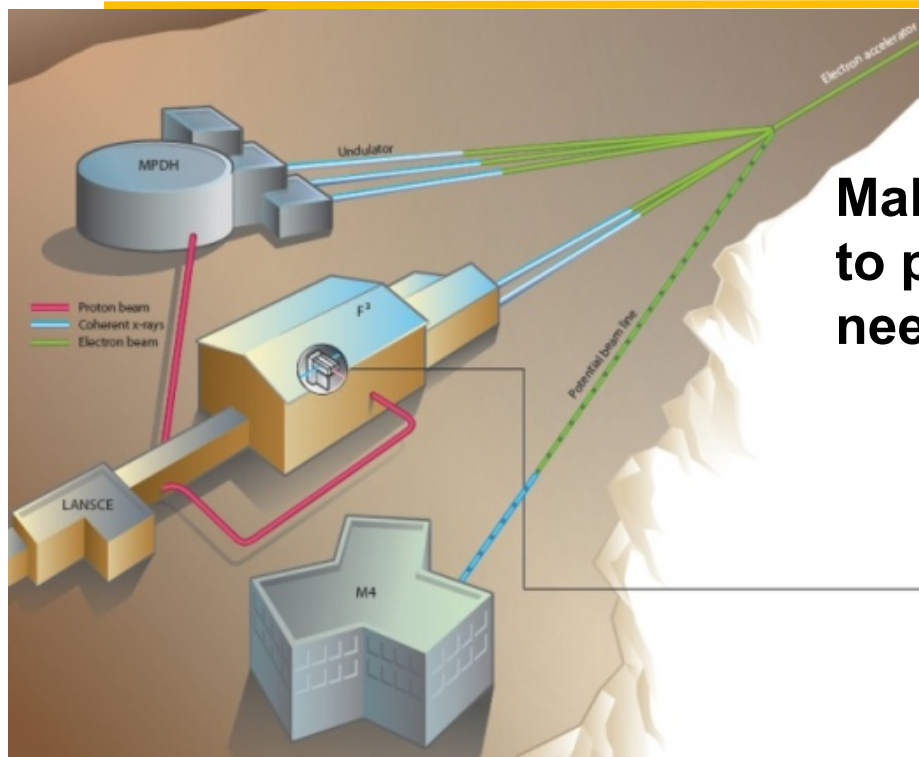


MaRIE photon needs can be met by an XFEL that is technically feasible and affordable

	MPDH	FFF		M4	
Energy/Range (keV)	50	50	50-600	< 5	5-50
Photons per image	10^{11}	10^{11}	10^9	10^9	10^{11}
Time scale for single image	50 fs	>1 s	0.001 s	50-500 fs	50 fs
Energy Bandwidth ($\Delta E/E$)	10^{-4}	10^{-4}	10^{-3}	10^{-4}	10^{-4}
Beam divergence	1 μ rad	1 μ rad	< 10 μ rad	< 10 μ rad	1 μ rad
Trans. coherence (TC) or spatial res.	TC	TC	1-100 μ m	TC	TC
Single pulse # of images/duration	100/1.5 μ s	-	-	-	-
Multiple pulse rep. rate/duration	120 Hz/day	0.01 Hz/mo.	0.01 Hz/mo.	1 KHz/day	0.01 Hz/days
Longitudinal coherence	yes	yes	no	no	yes
Polarization	linear	linear	no	Linear/circular	linear
Tunability in energy ($\Delta E/E/\text{time}$)	2%/pulse	fixed	fixed	10%/s	10x/day

- Photon energy - set by gr/cm² of sample and atomic number
- Photon number for an image - typically set by signal to noise in detector and size of detector
- Time scale for an image - fundamentally breaks down to transient phenomena, less than ps, and semi-steady state phenomena, seconds to months
- Bandwidth - set by resolution requirements in diffraction and/or imaging
- Beam divergence - set by photon number loss due to stand-off of source/detector or resolution loss in diffraction
- Source transverse size/transverse coherence - the source spot size will set the transverse spatial resolution, if transversely coherent then this limitation is not applicable so transverse coherence can be traded off with source spot size and photon number
- Number of images/rep rate/duration – images needed for single shot experiments/image rep rate/ duration of experiment on sample
- Repetition rate - how often full images are required
- Longitudinal coherence – 3D imaging
- Polarization - required for some measurements
- Tunability – time required to change the photon energy a fixed percentage

MaRIE will be the first capability with unique co-located tools necessary to realize transformational advances in materials performance in extremes



MaRIE builds on unique LANL capabilities to provide unique experimental tools needed to realize this vision:

In situ, dynamic measurements of real materials

Scattering & imaging simultaneously

in extreme environments

Dynamic & irradiation extremes

coupled to directed synthesis via predictive theory



Science-driven Requirements Lead to Integrated Facility Needs Fulfilled by MaRIE



Dynamic Extremes

Microstructure Evolution
Stochastic Explosive Microstructure & Detonation
Fluid/Mineral Interactions in
3-D Measurements of Turbulent

Radiation Extremes

Irradiation Stability of Structural Nanocomposites
Fission Gas Bubble & Swelling in UO₂ Nuclear Fuel
Mechanical Testing of Structural Materials in Fusion/Fission Environ.
Measurements of Temperature, Microstructure & Thermal Transport
Rad Damage in Passive Oxide Films & its Influence on Corrosion

Control of Complex Materials & Processes

Understanding Emergent Phenomena in Complex Materials
Developing Practical Superconductors by Design

Energy Conversion & Storage

Achieving Practical High-Density Energy Storage Through New Support/Catalyst Electrode Systems
Solar Energy Conversion w/ Functionally Integrated Nanostructures

Process-Aware Materials Performance

Nanostructured Ferritic Alloys
Exploring Separate Effects in Pu

Environments

Dynamic pressure <200 GPa
Strain rate = 10^1 – 10^7 s⁻¹
Temperature = 77–2000 K
High Explosives < 30 g
Pu isotope samples < 3 mm thick
Irradiation rate < 35 dpa/fpy
He(appm)/dpa ratios: 0.1-1, 9-13
Irrad Volume: 0.5 l @ >14 dpa/yr

Measurements

Scattering
Defects: 1 nm res over 10 μ m
Stress: 1-2 μ m res over 100 mm
Lattice Strain: 10 nm res in 3D
Density Imaging
0.1-1 nm, <1-ps res over 10 μ m
10 nm, <1-ps over 50 μ m
0.1-1 μ m, < 0.3 ns over 0.1-1 mm
Spectroscopic
3D chemistry mapping w/ 1 μ m res
Thermo-Physical Measurements
Temperature: 1 μ m res
Thermal Conductivity w/ 1 mW/m-K res

Synthesis with Characterization

Organic, inorganic, biomaterials incl nanomaterials, HE & actinides
Thin films with buried interface characterization

50 keV coherent x-ray source with 10^{11} photons per macropulse focused to 1-200 μ m

Dynamic charged particle imaging with 20-GeV electrons

Tunable ultrashort x-ray source for excitation: 5-35 keV, 100 fs, focused to 10 nm

Ultra short pulse lasers for spectroscopy: THz (2 meV) to VUV (6 eV)

MW fast neutron source with 2×10^{15} n/cm²-s and >4000 h/yr operation with < 10 beam trips per day over 1 min

Crystal growth with control of impurities & defects during and after fab

Deposition Lab w/CVD, PVD, evaporation, ion beams

Nanofabrication Lab w/ lithography, dry & wet etch, thermal processing

Characterization Lab w/ SEM, FE-SEM, AFM, SALVE, ion beams

Data Visualization Lab w/ 1MB-10TB available per expt.

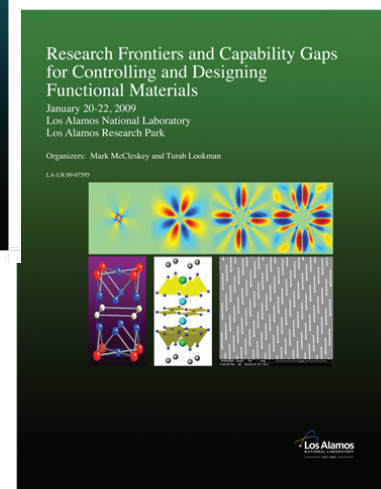
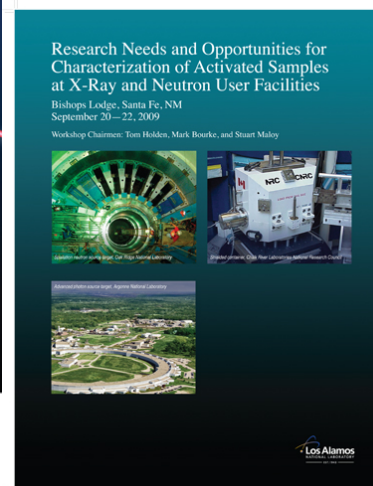
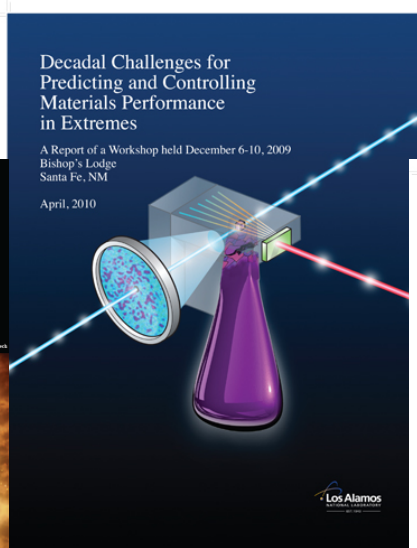
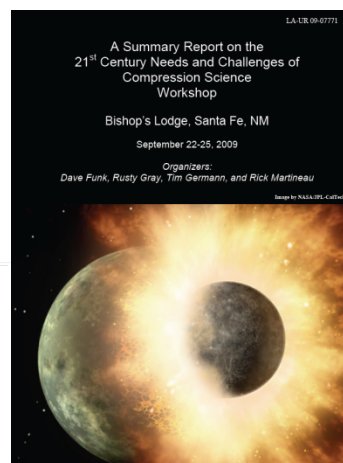
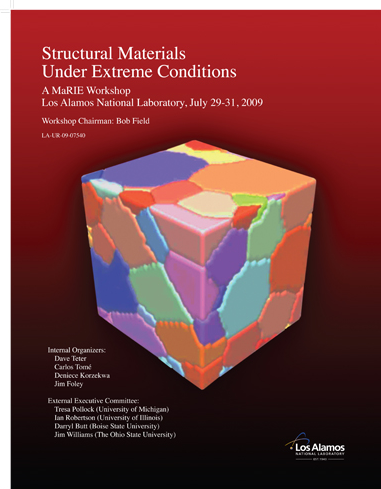


MaRIE builds upon existing \$B investments at LANSCE with the addition of the:

- Electron Linac with XFEL Systems
- Multiprobe Diagnostic Hall
- Fission-Fusion Materials Facility
- Making, Measuring, & Modeling Material Facility

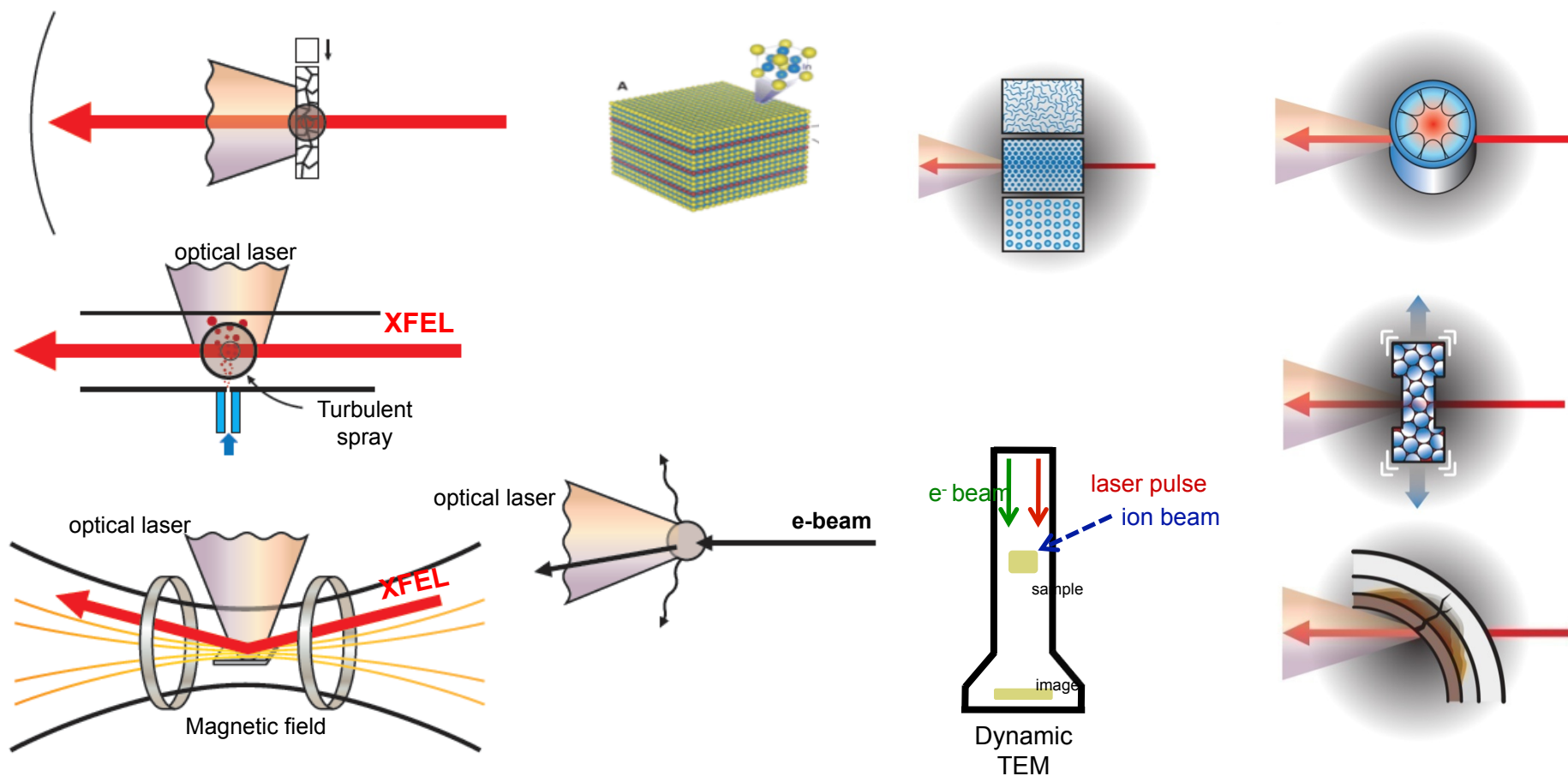


Community-based workshops have helped to define the decadal challenges for predicting and controlling materials performance in extremes





First experiment teams include ~170 scientists from ~ 60 institutions in 10 countries



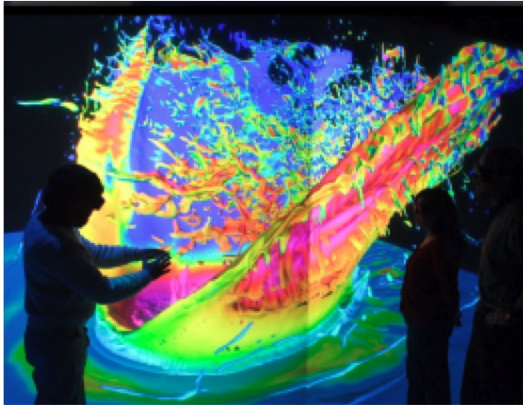
MaRIE will address problems central to Department of Energy missions in energy, science, and security



- **What are the consequences of materials failure for weapons performance?**
- **How do we accelerate the certification of materials to enable a nuclear renaissance?**
- **Can we predict and prevent materials damage?**
- **Can we discover by design materials to perform in unprecedented irradiation extremes?**
- **How do we predict and control microstructure for designed materials performance?**
- **Can we design and synthesize new materials with controlled functionality?**

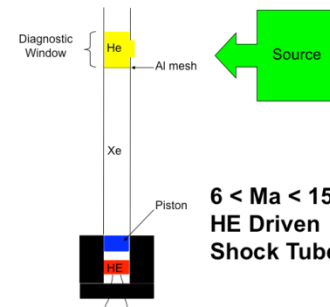
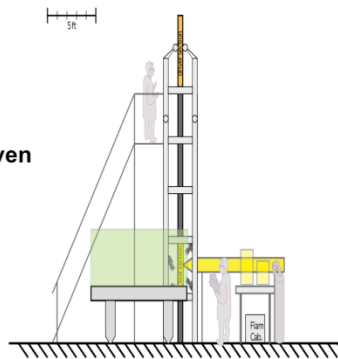
Example: Performance after material “failure”

Developing predictive capability across all relevant scales for turbulent flows, including those with “strength”



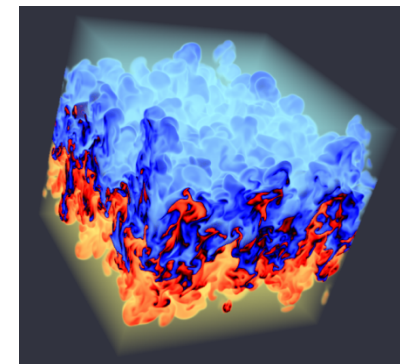
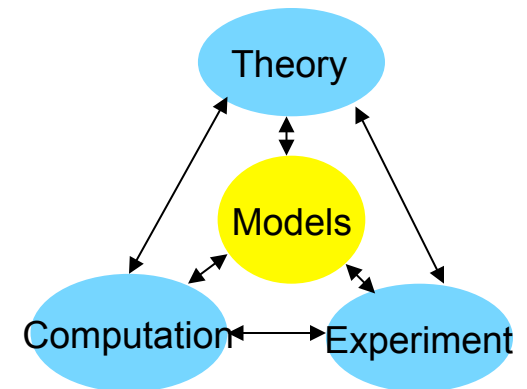
The goal :- Develop predictive capability for turbulent mix

**0 < Ma < 6
Pressure Driven
Shock Tube**



**6 < Ma < 15
HE Driven
Shock Tube**

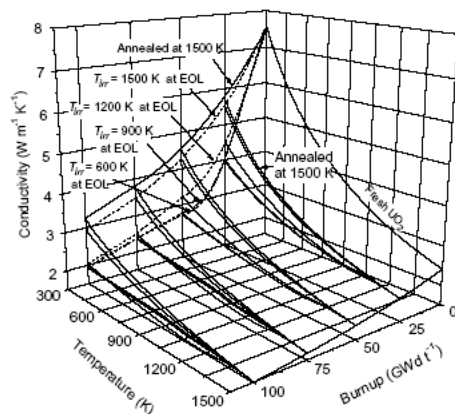
The first experiment :-Multi-scale fluid dynamics experiments with the ability to measure turbulent flows at all relevant space and time scales (μm and μsec), featuring opaque materials and/or high-velocity flows requiring high repetition measurements.



The model :-Direct Numerical Simulation coupled to Reynolds-Averaged Navier Stokes turbulence model



Determining spatially resolved thermophysical properties in prototype fuel geometries



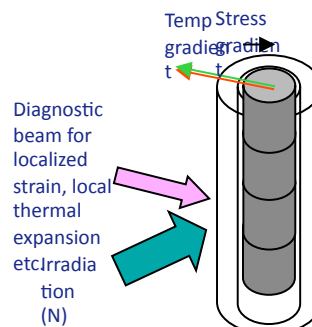
Predicted and measured UO₂ thermal conductivity

Goal :- Spatially resolved predictions and measurements of engineering performance of prototype fuel pin geometries as a function of power, burnup and time

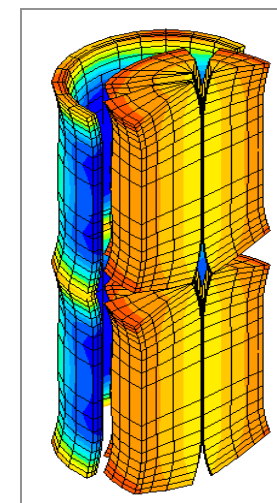


Team includes: Marius Stan et al. (LANL, ANL, Wisconsin, INL, CEA ...)

Operated by Los Alamos National Security, LLC for NNSA



Experiment :- MaRIE will use photons, (electrons & neutrons) to make unique measurements of phase, strain, microstructure, porosity & temperature distributions on engineering scale samples in & out of a radiation environment



(PLEIADES code, CEA, France)

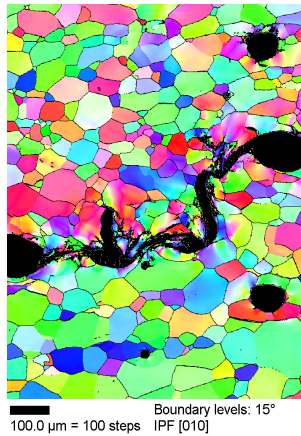
Model :- Stress/ Temperature Field in a Fuel Element consisting of two ceramic pellets and metallic clad.

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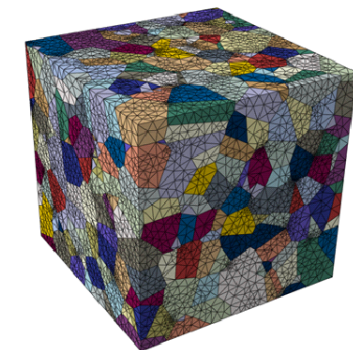
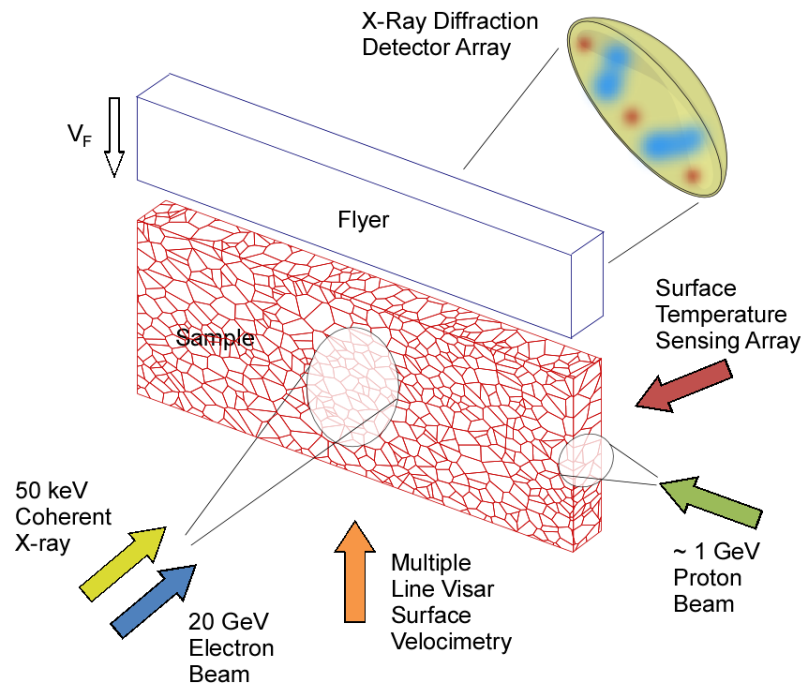




Understanding the role of microstructure-based heterogeneity evolution in material damage



The goal :- Predict dynamic microstructure and damage evolution



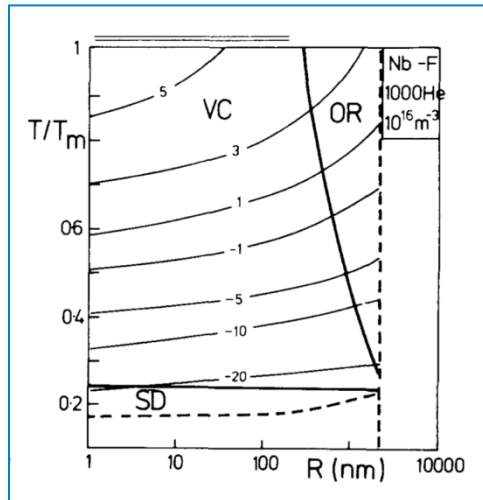
Meso-Scale Theory and Simulation

The first experiment :- Multiple, simultaneous dynamic in situ diagnostics with resolution at the scale of nucleation sites ($< 1 \mu\text{m}$; ps – ns)

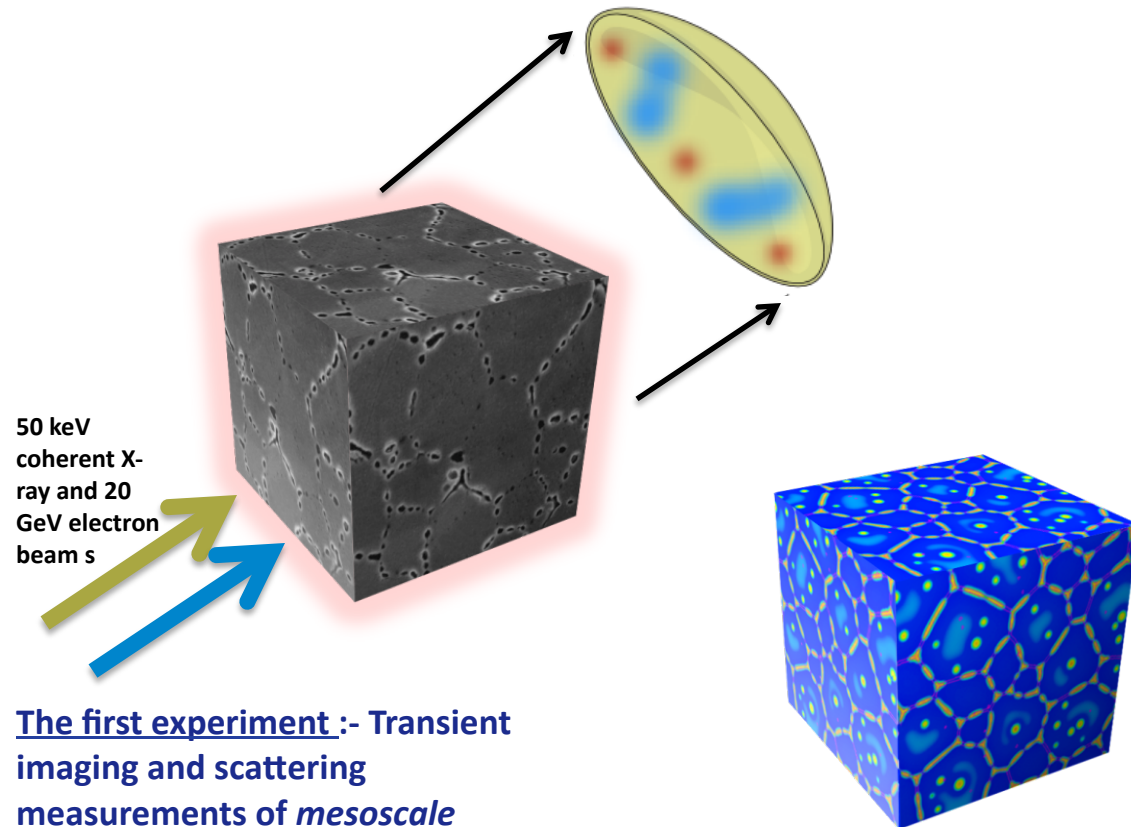
The model :- Accurate sub-grain models of microstructure evolution coupled to molecular dynamics



Understanding creep via transient measurements of cavity growth under fast neutron irradiation



The goal :- Predicted and measured cavity growth mechanism maps of creep under extreme irradiation conditions

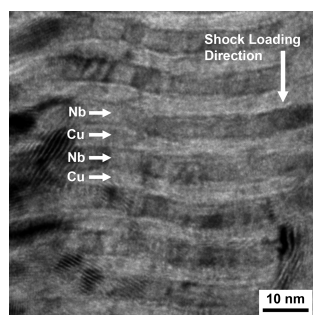


The first experiment :- Transient imaging and scattering measurements of *mesoscale* objects with *nm spatial resolution* in an extreme radiation environment

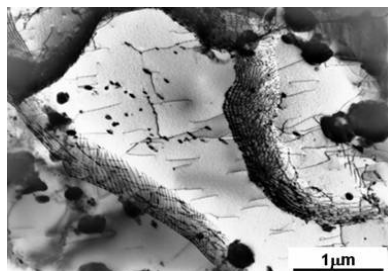
The model :- Phase field simulation of heterogeneous nucleation of gas bubbles



Understanding the role of interfaces in strain evolution

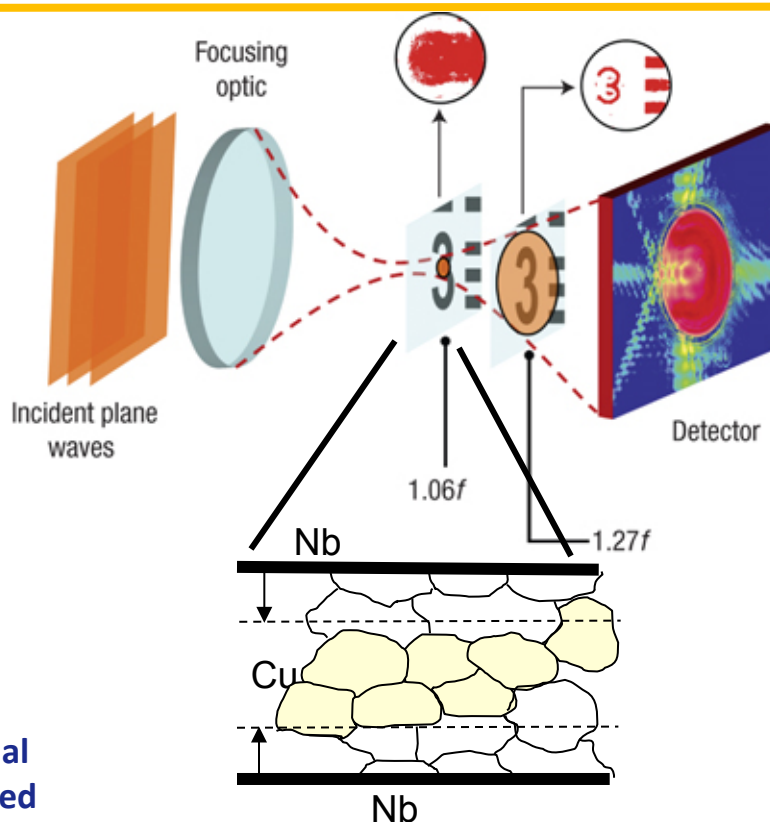


Nano laminates

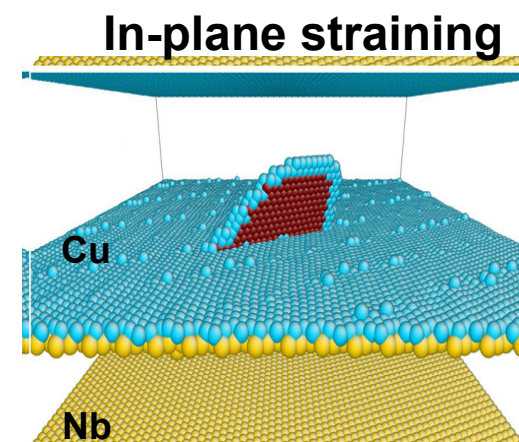


ODS steel

The goal : Predict interfacial microstructure for increased strength and irradiation resistance



The first experiment :
3-D movies of dislocation dynamics in materials at buried interfaces, micron field of view with focusing at nm resolution

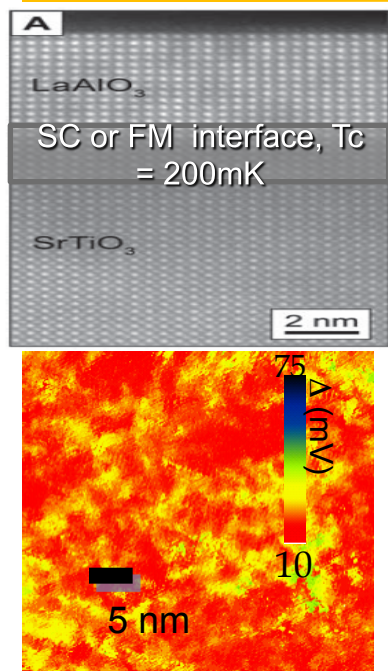


The model : Advanced M²S with micron scale, multigranular predictions

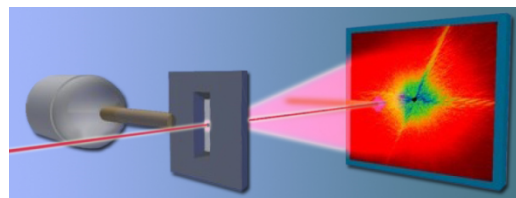
Example: Design and synthesis of new materials with controlled functionality



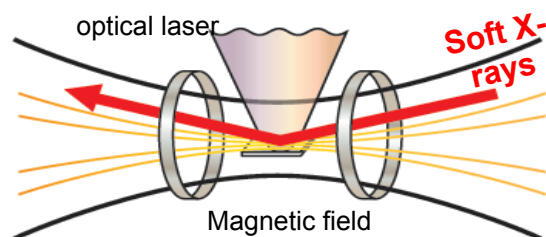
Understanding complex functionality beyond Bloch & Boltzmann



The goal :- Predict and control functionality at interfaces with complex energy landscapes



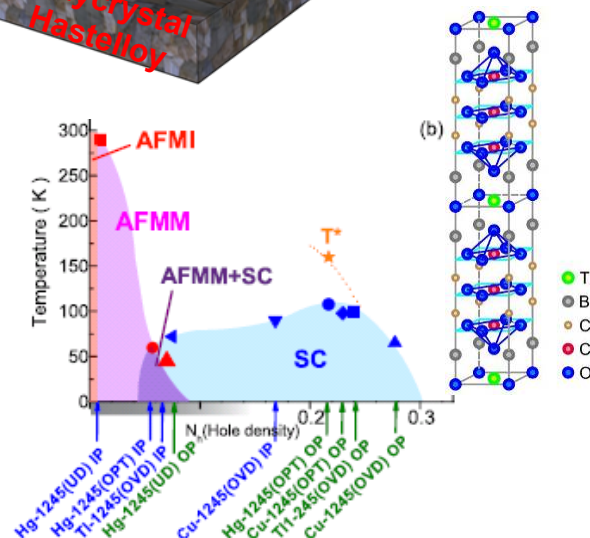
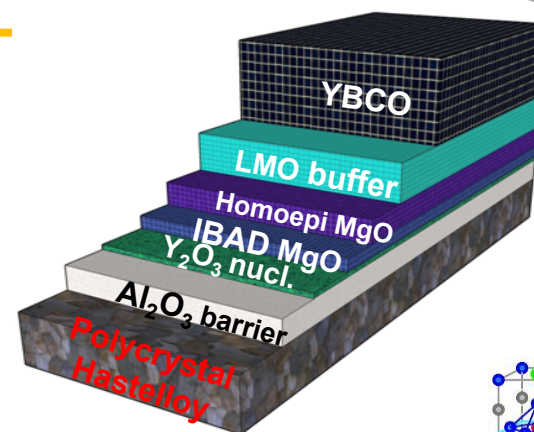
High energy diffraction



Soft energy electronic and magnetic information

The first experiment

Visualization overlay and comparison of ultrafast 3-D imaging of structure using hard X-rays and ultrafast 3-D imaging of functionality with soft X-rays

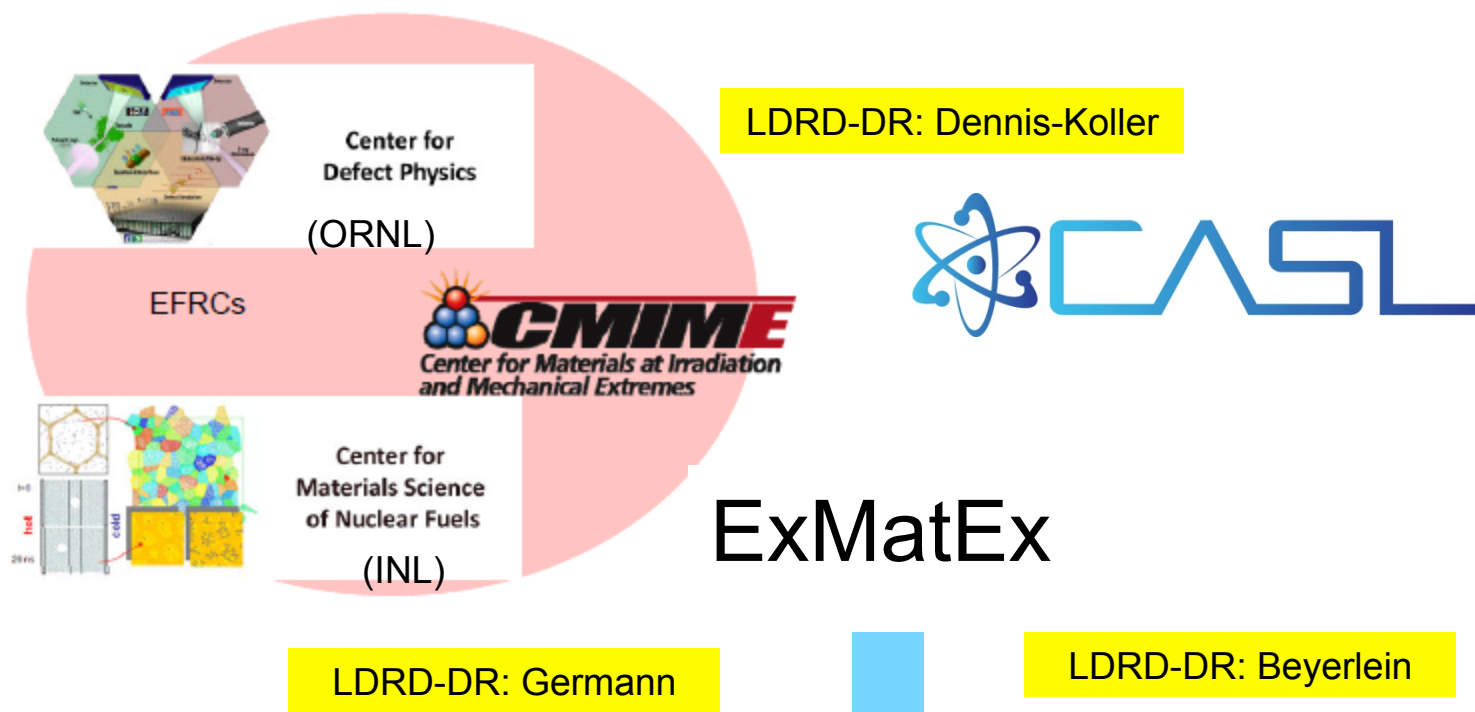


The model : Beyond periodic repetition of structure (interfaces), beyond temporal limits of Boltzmann

Example: Doing aspects of MaRIE science today



Recent efforts to integrate theory and experiment through co-design for materials in extremes are succeeding



DOE "Blue Skies" Initiatives

Materials in Extremes/Irradiation Resistant Materials (BES/FES/ASCR)/NNSA/NE

Exascale Computing - ASCR/NNSA

OSTP: Computational Materials by Design for Innovation



Operated by Los Alamos National Security, LLC for NNSA

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Slide 37



Example: Peer facilities for accelerating certification and predicting damage



Current capabilities enable collaborations, help refine MaRIE facility requirements, and enable science exploration

Controlled Functionality; Process-Aware Certification; Transformational Performance

and BEYOND

DECADE

MPDH

F³

M4

pRad @ GSI

LCLS-MEC

DC-CAT

MTS

HFIR, ATR

JANNuS

Exascale Co-design

Nanocenters

EFRCs

PRESENT

CAPABILITIES

p, e-
imaging

X-ray
Sources

Lasers,
Drivers

In-Situ
Measurements

Relevant
Doses

Lab-Scale
Extremes

In-Situ
Char.

Synthesis
Capability

Multi-Probe
Measurements

Extreme
Environments

Directed
Synthesis

Integration
(esp. theory)

Control
↑
Observation



At LANSCE today, a flexible 1 MW, 800 MeV proton accelerator drives several user facilities



Unique, highly-flexible beam delivery to multiple facilities 6 mo/yr @ 24/7 with ~ 1200 user visits

Lujan Center

- *Materials science and condensed matter research*
- *Bio-science*
- *Nuclear physics*
- *A National BES user facility*

WNR

- *Nuclear physics*
- *Semiconductor irradiation*

Ultra-cold Neutron Facility

- *Fundamental nuclear physics*

Proton Radiography

- *HE science, dynamic materials science, hydrodynamics*

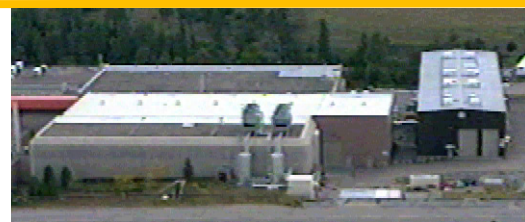
Isotope Production Facility

- *Nuclear medicine*
- *Research isotope production*

LANL National User Facilities form a synergistic triad for materials research



Nano-materials
synthesis and
characterization

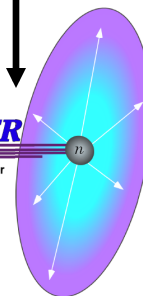


Research with high
magnetic fields

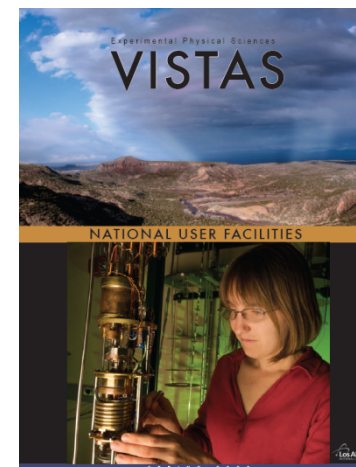


LUJAN CENTER

Los Alamos Neutron Science Center
Los Alamos National Laboratory



Neutron
scattering





User opportunities exist today for “MaRIE relevant” measurements at the Lujan Center

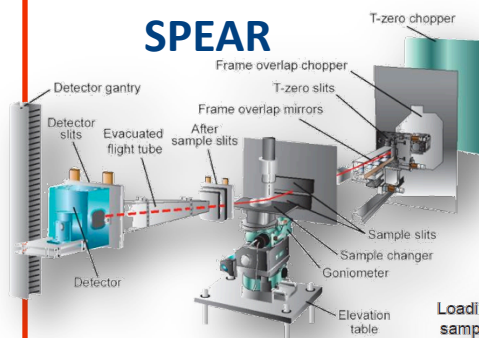
Sample

*Pb-lined
Chalk River
“Castle”
Scattering
solid angle 80
degrees*

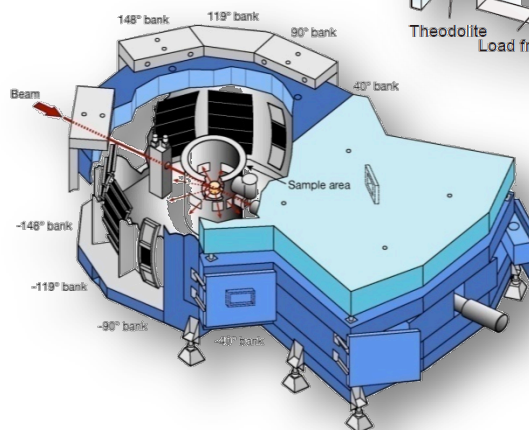
*RADSPEC
instrument*



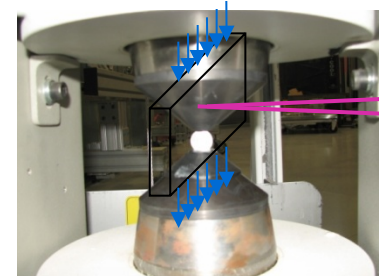
**Corrosion
SPEAR**



**Local structure
NPDF**

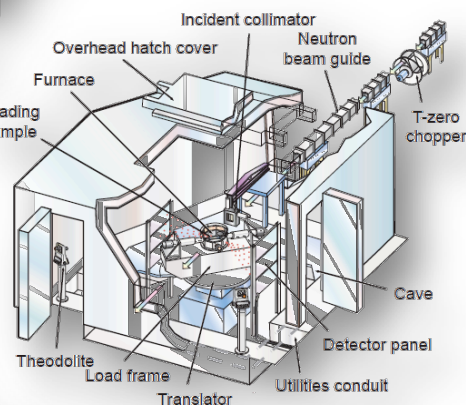


Magnetic field

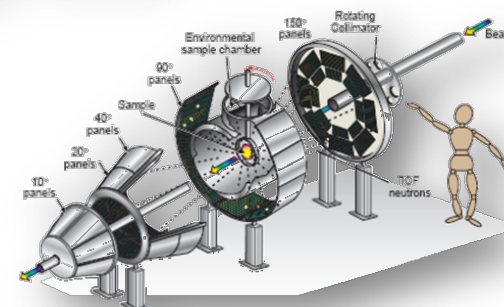


**Magnetic
Contrast
LQD**

**Stress and
Temperature
SMARTS**

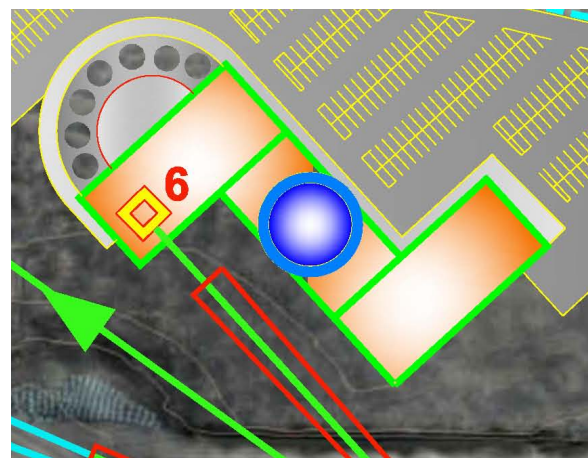
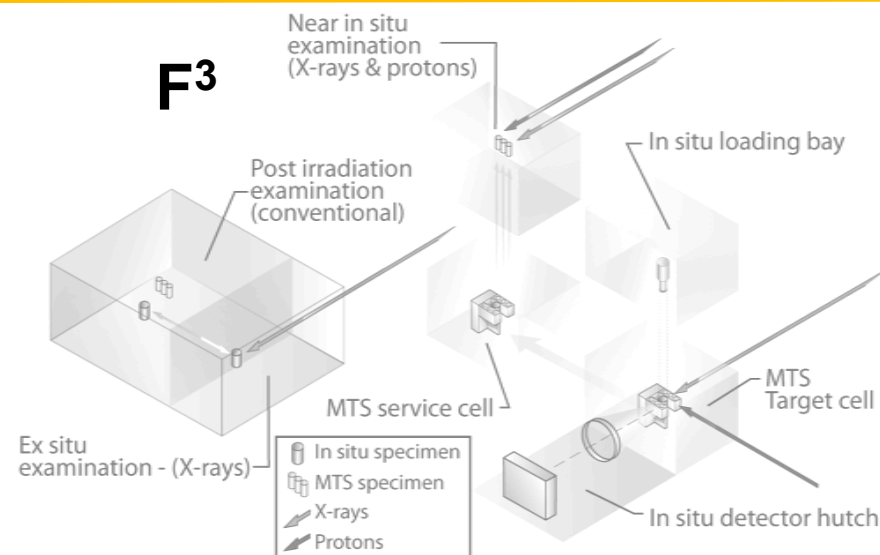
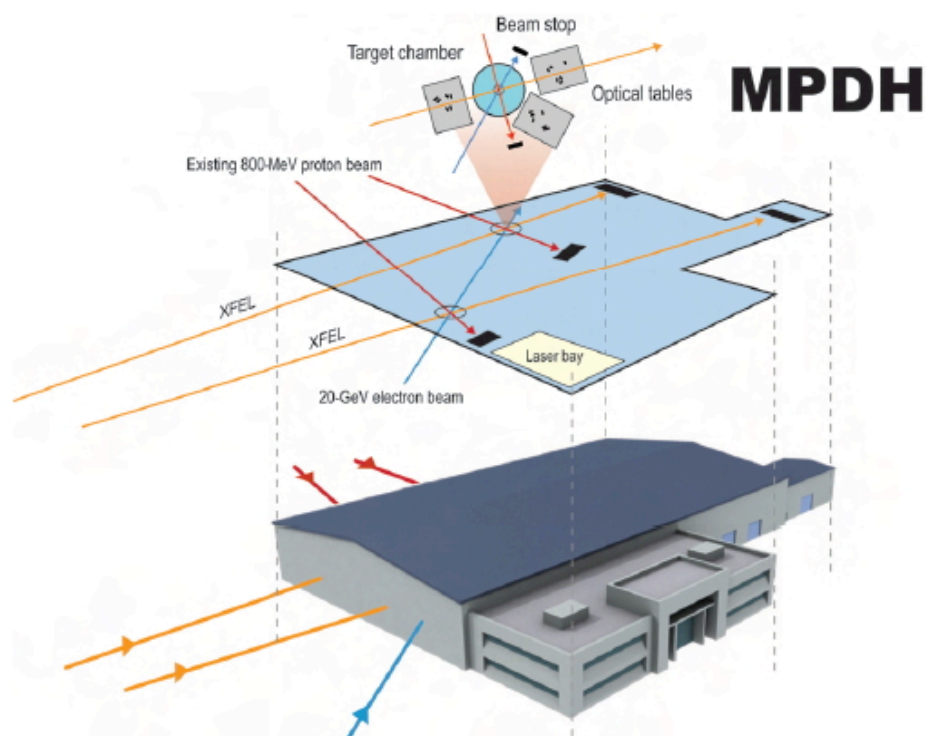


**Texture
HIPPO**



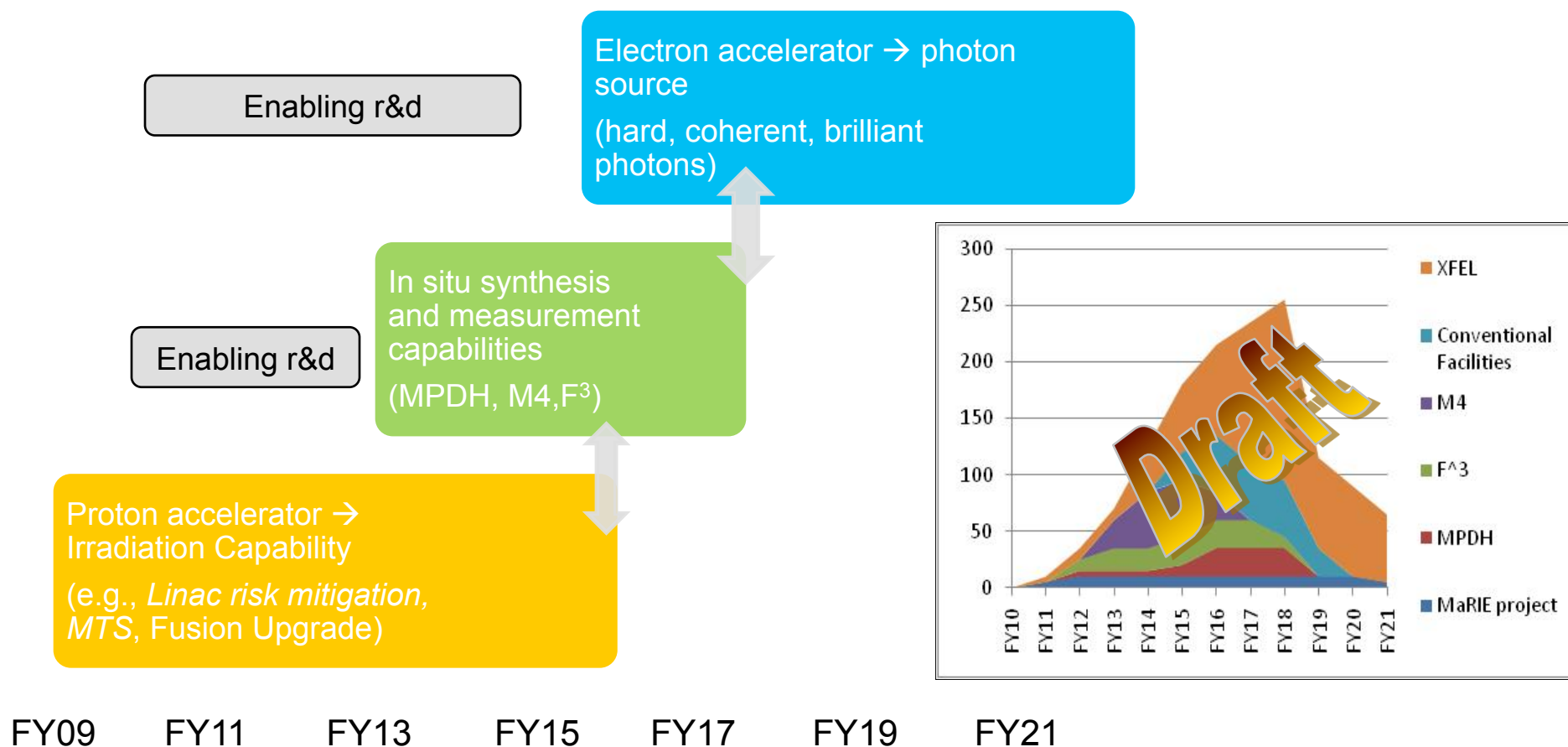


Preconceptual facility designs enable cost/risk/benefit analysis and cost estimates





MaRIE capabilities can be realized through a phased approach





MaRIE will be the first capability with unique co-located tools necessary to revolutionize materials in extremes

- A mission need exists for prediction and control of materials in extreme environments
- MaRIE will provide simultaneous in situ, transient measurements on real materials in relevant extremes coupled to directed synthesis and characterization through predictive theory
- Building on existing capabilities at LANL, MaRIE provides unprecedented international user resources
- MaRIE facility definition is being driven by community-validated performance gaps & functional requirements

